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Danish Atomic Energy Commission
Research Establishment Risø

Interpretation of Absolute Measurements of Radioactive Source Strength by the 4π Beta-Gamma-Coincidence Method

by J. Thomas



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by

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Abstract

The present report deals with the interpretation of absolute measurements of radioactive source strength by the 4π beta-gamma-coincidence method. After a description of the statistical behaviour of a generalized multiscaler instrument, the special case of the beta-gamma-coincidence method is evaluated.

The theory is demonstrated by the measurement of Au^{198} in gold foils, where the final accuracy is shown to be between 0.1 and 0.2 per cent.

Special attention is given to the count-rate-dependent corrections, which are here measured directly by means of the substitution method.

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Contents

	Page
Chapter 1: Introduction	5
Chapter 2: The Statistical Behaviour of Counting Systems in General	8
Chapter 3: The Probability Distribution of the Parameters	13
Chapter 4: The Beta-Gamma-Coincidence Method	19
Evaluation of the Integral in the Denominator	27
Some Figures Characterizing the Function (4.38) ...	31
Chapter 5: Description of the Actual Experiment	36
The Decay of Au ¹⁹⁸	36
The Beta Detector	38
The Gamma Detector	41
The Electronics and the Coincidence Unit	44
Corrections for Finite Foil Thickness	46
Comparison with the He ³ -Counter	54
Chapter 6: Count-Rate-Dependent Corrections	59
The Substitution Method in the Case of Au ¹⁹⁸	63
Subtraction of Background	64
The Half-Life of the Foil Activity	71
Chapter 7: Conclusion	77
Acknowledgements	79
References	80
Errata to Risø Report No. 70	85

CHAPTER 1: INTRODUCTION

During the first ten years of this century a great effort was made to establish the nature of radioactive decay and the radiation from radionuclides. The status in 1904 was excellently reviewed by M^{me} S. Curie (1904), and E. Rutherford (1907) reviewed the method of observing the emission by its action on photographic plates, by its ionization in gases and by scintillation in different materials.

The question of the constancy of a long-lived radioactive source was confused by observations of time fluctuations by H. L. Bronson, who observed slight oscillations of the electrometer needle connected to an ionization chamber, and claimed, "The most probable explanation seems to be that it is due to exceedingly small and rapid changes in the ionization itself" (H. L. Bronson, 1906). At the same time E. v. Schweidler (1905) published a statistical explanation of the time fluctuations in the number of decays in a radioactive source. Experiments on the statistics were made in the following years (K. W. F. Kohlrauch, 1906; E. Meyer and E. Regener, 1908), but an absolute determination of the number of emitted α -particles from a radioactive source by observation of the irregularities in an ionization-chamber current (H. Geiger, 1908) was only possible after the establishment of the charge of an α -particle (E. Rutherford, 1906 b). Even with this limitation, the first absolute measurement of the number of α -particles emitted from radium was made by E. Rutherford (1905) by measuring the total electrical charge carried by the α -particles in vacuum. It was also demonstrated that the directions of the emitted α -particles were on an average uniformly distributed if the radioactive layer was very thin (E. Rutherford, 1906 a).

The efficiency of the scintillating zinc-sulphide screen was determined to be between 96 and 99 per cent (E. Regener, 1908; E. Rutherford and H. Geiger, 1908), and the problem of main interest in the absolute determination of the number of α -particles emitted from e. g. radium was the question of the decay constant λ of radium. Because of the long half-life of radium it was impossible to observe experimentally any decay in the activity of an enclosed sample, and the decay constant had to be found from equation (1.1):

$$-\frac{dN}{dt} = \lambda N, \quad (1.1)$$

where N is the number of radioactive atoms in the sample and $-dN/dt$ the number of decays in a unit time interval.

The constancy of the decay constant was very important, and, except for a few special decay types (orbital electron capture and isomeric transitions), the decay constant of a radionuclide is unaffected by temperature, pressure, concentration of the sample, chemical composition, electric and magnetic fields, etc.. In a modern theory referred to as the theory of "Hidden Variables" (Winter, 1962) it is claimed that the decay constant may change as a function of the time in which the radionuclide exists. In experiments with radium emanation (Em-22) M^{me} P. Curie (1910) measured over 11 half-lives, and Rutherford and Tuomikoski (Rutherford, 1911) measured over 27 half-lives, and no divergence from an exponential decay was observed. In connection with a recent measurement of Mn^{56} over 34 half-lives where no divergence from exponential decay was observed either, Rolf G. Winter stated, "At present, we can conclude that "hidden variables" are hidden very well" (R. G. Winter, 1962).

The statistical nature of the radioactive decay was studied by E. Rutherford and H. Geiger (1910), M^{me} P. Curie (1911), and E. Marsden and Baratt (1911), and all experiments were explained perfectly by a theory given by H. Bateman (1910) on the basis of the earlier publication by E. v. Schweidler (1905). A detailed calculation on a series of classical experiments was given by L. v. Brotkiewicz (1913) and later by A. E. Ruark and L. DeVol (1935 and 1936). In all these statistical treatments the assumption was that the activity and other parameters of the experiments were known and the mean value and spread of the observations could be derived from them. No attempts were made to find the values of the parameters, which in most cases are unknown, on the basis of the observations. This problem was first mentioned by the Reverend Thomas Bayes as early as 1763 (T. Bayes, 1763) and has later been touched upon by L. J. Rainwater and C. S. Wu (1947), who give the inverse probability distribution in the case of Poisson distribution, but without any statement of the method of derivation. In refs. II and III the author generalizes the postulate by T. Bayes (1763), and in the present report the problem of deriving the inverse probability in the case of a coincidence measurement is solved.

Coincidence measurements were first made by H. Geiger and E. Marsden (1910) in an investigation of radioactive series decay, and coincidence techniques were first applied to the problem of absolute counting by H. Geiger and A. Werner (H. Geiger, 1924; H. Geiger and A. Werner, 1924),

in both cases with visual scintillations. The two observers were each equipped with a telegraphic key, and the signals herefrom were recorded by a three-pen telegraph recorder (the third pen was used to record time signals). A few years later W. Bothe (1929) reported an electronic instrument in which the coincidences were identified directly on the electronic pulses before registration; since then this technique has been used.

The special advantage of using a 4π proportional counter for the β -particles in β - γ -coincidence counting, and an absorber to ensure that no β -particles should enter the gamma detector, was pointed out by W. Bothe and H. J. v. Baeyer (1935). A review of the applications of the coincidence techniques has been given by J. V. Dunworth (1940).

In connection with the great production of artificial radionucleides after the invention of the nuclear reactor the interest in absolute measurements was revived (J. L. Putman, 1950), and the proceedings from a series of international conferences (Washington, 1949; Zurich, 1957; Tidewater Inn, 1957; Vienna, 1959) show the later developments in the methods and the subjects.

From time to time the units of radioactivity have changed, and at present the curie unit is just an abbreviation for the rate

$$1 \text{ Ci} = 3.7 \cdot 10^{10} \text{ per second} \quad (1.2)$$

(NBS Handbook 84, 1962).

In the following chapters the theory of inverse probability is used in the interpretation of a 4π β - γ -coincidence experiment. The point of actual interest is the absolute disintegration rate of samples of the radionucleide Au^{198} used in the determination of the neutron density in a neutron beam, and the results of the method are compared with standardizations of the same beam by means of a He^3 -filled proportional counter (J. Als-Nielsen, A. Bahnsen and W. K. Brown, 1966). When the neutron density in the beam is determined in this way, a measurement of the number of decays of neutrons in a known volume of the beam will yield the decay constant of the neutron according to formula (1.1) (C. J. Christensen, A. Nielsen, A. Bahnsen, W. K. Brown, and B. M. Rustad, 1966).

A special part of the work has already been published and is referred to as refs. I, II and III, standing at the beginning of the reference list.

CHAPTER 2: THE STATISTICAL BEHAVIOUR OF COUNTING SYSTEMS IN GENERAL

Before we shall look at the special problems concerning the α - β - γ - coincidence instrument, it is worth while to investigate counting systems on a more general basis. We therefore regard a nuclear instrument with a radioactive source A , in which the probability of decays is a function of space and time. Let us say that the probability that a decay will occur in the volume element $dx dy dz$ around the point (x, y, z) and in the time element dt after the time t is given as

$$A(x, y, z, t) dx dy dz dt. \quad (2.1)$$

Some of these decays are observed by means of nuclear detectors of some kind, and the numbers of observed decays are registered in s scalers. Let us denote the probability that a decay occurring in the point (x, y, z) at the time t will be registered in the i^{th} scaler by

$$\epsilon_i(x, y, z, t). \quad (2.2)$$

Let us assume that the probabilities of the decay and of the registration are independent of each other. Then the probability of the registration in the i^{th} scaler of a decay taking place in the volume-time element $dx dy dz dt$ will be

$$\epsilon_i(x, y, z, t) A(x, y, z, t) dx dy dz dt. \quad (2.3)$$

If we further assume that the probability of a decay occurring and being registered is independent of the occurrence of any other decay (whether registered or not), then the probability G of obtaining n_i' registrations in the i^{th} scaler of decays in the volume element $dx dy dz$ during the time from 0 to t' will be given by the Poisson distribution

$$G(\epsilon_i(x, y, z, t), A(x, y, z, t), 0, t'; n_i') = \frac{(p_i')^{n_i'}}{n_i'!} e^{-p_i'}, \quad (2.4)$$

where

$$P_i' = \int_0^{t'} \epsilon_i(x, y, z, t) A(x, y, z, t) dt \quad (2.5)$$

(Ruarh and Devol, 1936).

If we now take the individual volume elements in the radioactive source as independent sources, we can use the additive nature of radioactive source strengths, as shown in ref. II, and we find the probability of registering - in the i^{th} scaler - the total number n_i of decays from all parts of the source during the time from 0 to t' to be

$$G(\epsilon_i(x, y, z, t), A(x, y, z, t), 0, t'; n_i) = \frac{P_i^{n_i}}{n_i!} e^{-P_i}, \quad (2.6)$$

where

$$P_i = \int_{\Omega} \left\{ \int_0^{t'} \epsilon_i(x, y, z, t) A(x, y, z, t) dt \right\} dx dy dz. \quad (2.7)$$

Ω indicates that the integration should be carried out over the total volume of the radioactive source.

Let us again for a while look at the small volume $dx dy dz$ around (x, y, z) and, for the sake of paper economy, write

$$A(x, y, z, t) dx dy dz dt = A(t) dt \quad (2.8)$$

and

$$\epsilon_i(x, y, z, t) = \epsilon_i(t). \quad (2.9)$$

Let us now assume that registration of a decay in one scaler excludes the registration of the same decay in the other scalars, and that

$$\sum_{i=1}^S \epsilon_i(x, y, z, t) = 1 \quad (2.10)$$

for all (x, y, z, t) .

we will now ask for the joint probability of obtaining the set of registrations (n_1, \dots, n_s) in the s scalars during the time from 0 to t' ; let us denote this probability

$$\begin{aligned} G(\epsilon_1(t), \dots, \epsilon_s(t), A(t), 0, t'; n_1, \dots, n_s) \\ = G(t'; n_1, \dots, n_s), \end{aligned} \quad (2.11)$$

where the last notation is shorthand.

The probability of obtaining the set of (n_1, \dots, n_s) registrations in the time from 0 to $t' + dt$ may now be written

$$\begin{aligned} G(t' + dt; n_1, \dots, n_s) \\ = G(t'; n_1, \dots, n_s) (1 - A(t) dt) \\ + A(t) dt \{ G(t'; n_1 - 1, \dots, n_s) \epsilon_1(t) + \dots + G(t'; n_1, \dots, n_s - 1) \epsilon_s(t) \} \end{aligned} \quad (2.12)$$

since $A(t)dt$ is the probability of one decay in the small time interval dt .

By a little rearrangement we find

$$\begin{aligned} \frac{G(t' + dt; n_1, \dots, n_s) - G(t'; n_1, \dots, n_s)}{dt} \\ = \frac{d}{dt} G(t'; n_1, \dots, n_s) \\ = -G(t'; n_1, \dots, n_s) A(t) \\ + G(t'; n_1 - 1, \dots, n_s) A(t) \epsilon_1(t) + \dots + G(t'; n_1, \dots, n_s - 1) A(t) \epsilon_s(t), \end{aligned} \quad (2.13)$$

which is a differential equation with the solution

$$G(t'; n_1, \dots, n_s) = \frac{\left(\int_0^{t'} A(t) \epsilon_1(t) dt \right)^{n_1}}{n_1!} \dots \frac{\left(\int_0^{t'} A(t) \epsilon_s(t) dt \right)^{n_s}}{n_s!} e^{-\int_0^{t'} A(t) dt} \quad (2.14)$$

This can be proved by differentiating (2.14). From the assumption (2.10) we find

$$\begin{aligned} \int_0^{t'} A(t) dt &= \int_0^{t'} \sum_{i=1}^s \varepsilon_i(t) A(t) dt \\ &= \sum_{i=1}^s \int_0^{t'} \varepsilon_i(t) A(t) dt, \end{aligned} \quad (2.15)$$

and using this, we obtain in the complete notation

$$\begin{aligned} G(\varepsilon_1(t), \dots, \varepsilon_s(t), A(t), 0, t'; n_1, \dots, n_s) \\ = \frac{\left(\int_0^{t'} A(t) \varepsilon_1(t) dt \right)^{n_1}}{n_1!} e^{-\int_0^{t'} A(t) \varepsilon_1(t) dt} \\ \dots \frac{\left(\int_0^{t'} A(t) \varepsilon_s(t) dt \right)^{n_s}}{n_s!} e^{-\int_0^{t'} A(t) \varepsilon_s(t) dt} \end{aligned} \quad (2.16)$$

which is the product of s Poisson distributions.

The integration over the whole source volume is again obtained by means of the above-mentioned additivity of source strengths. We find

$$\begin{aligned} G(\varepsilon_1(x, y, z, t), \dots, \varepsilon_s(x, y, z, t), A(x, y, z, t), 0, t'; n_1, \dots, n_s) \\ = \prod_{i=1}^s \frac{\left(\iiint_{\Omega} dx dy dz \int_0^{t'} \varepsilon_i(x, y, z, t) A(x, y, z, t) dt \right)^{n_i}}{n_i!} \\ e^{-\iiint_{\Omega} dx dy dz \int_0^{t'} \varepsilon_i(x, y, z, t) A(x, y, z, t) dt}, \end{aligned} \quad (2.17)$$

where n_i is now the total number of registrations in the i^{th} scaler.

The joint distribution just found (equation 2.17) is seen to be the product of the distribution functions for the numbers of registrations in the individual scalers as obtained by means of equations (2.6) and (2.7). Thus it is proved that the numbers of registrations in the s scalers are independent statistical variables.

If we multiply numerator and denominator in formula (2.17) by

$$(n_1 + \dots + n_s)! \left(\iiint_{\Omega} dx dy dz \int_0^{t'} A(x, y, z, t) dt \right)^{n_1 + \dots + n_s}, \quad (2.18)$$

we obtain

$$\begin{aligned} G(\epsilon_1(x, y, z, t), \dots, \epsilon_s(x, y, z, t), A(x, y, z, t), 0, t'; n_1, \dots, n_s) \\ = \frac{a^{n_1 + \dots + n_s}}{(n_1 + \dots + n_s)!} e^{-a} \frac{(n_1 + \dots + n_s)!}{n_1! \dots n_s!} q_1^{n_1} \dots q_s^{n_s}, \end{aligned} \quad (2.19)$$

where

$$a = \iiint_{\Omega} dx dy dz \int_0^{t'} A(x, y, z, t) dt \quad (2.20)$$

and

$$q_i = \frac{\iiint_{\Omega} dx dy dz \int_0^{t'} \epsilon_i(x, y, z, t) A(x, y, z, t) dt}{\iiint_{\Omega} dx dy dz \int_0^{t'} A(x, y, z, t) dt} \quad (2.21)$$

Equation (2.19) shows that the joint distribution may be regarded as a Poisson distribution for the total number of decays during the time from 0 to t' from an activity, a , multiplied by a multinomial distribution describing the distribution of the said total number in the s different and independent groups.

The only assumption necessary for the above treatment was that both the decays and the registrations were independent of any other decay or registration, and thus the result is only applicable in the limit of very low count-rates, where effects such as resolving and paralysis times are without importance. These problems will be treated in the chapter "Count-Rate-Dependent Corrections".

CHAPTER 3:

THE PROBABILITY DISTRIBUTION OF THE PARAMETERS

Let us assume that we have s independent observations of numbers of occurrences of some specified events (e. g. registrations of radioactive decays), and let us assume that the counting of each type of events is governed by a Poisson probability distribution. Thus the probability of observing n_i of the i^{th} type of events is given by

$$G(p_i; n_i) = \frac{p_i^{n_i}}{n_i!} e^{-p_i} \quad (3.1)$$

where p_i is the parameter characteristic of the i^{th} type of events.

The joint distribution giving the probability of observing the set of counts (n_1, \dots, n_s) is, according to the assumed independency,

$$G(p_1, \dots, p_s; n_1, \dots, n_s) = \frac{p_1^{n_1}}{n_1!} e^{-p_1} \dots \frac{p_s^{n_s}}{n_s!} e^{-p_s} \quad (3.2)$$

$$= \frac{a^n}{n!} e^{-a} \frac{n!}{n_1! \dots n_s!} q_1^{n_1} \dots q_s^{n_s}, \quad (3.3)$$

where

$$n = n_1 + \dots + n_s, \quad (3.4)$$

$$q_i = \frac{p_i}{p_1 + \dots + p_s}, \quad (3.5)$$

and

$$a = p_1 + \dots + p_s \quad (3.6)$$

with the connection

$$q_1 + \dots + q_s = 1 \quad (3.7)$$

between the q 's.

It is not necessary in the derivation of equation (3.2) to impose any limitation on the values of the parameters p_i except that they should all be positive. They may be connected by some specified relation, or they may all be completely independent without violating the assumption that the observation of numbers of occurrences is independent. An example of correlation between the parameters is the mere repetition of the observations of the same type of events where all the p 's have the same value and where the observation of the numbers n_i can still be made independently.

As in the case with a single parameter (ref. II), we can now introduce the a priori probability

$$P(p_i) dp_i \quad (3.8)$$

that p_i lies in the interval from p_i to $p_i + dp_i$ independently of the observation of any n_i . It may be that the joint probability

$$P(p_1, \dots, p_s) dp_1 \dots dp_s \quad (3.9)$$

cannot be expressed as the product of the individual probabilities (3.8) if there exists information on relations between the p 's.

With the a priori probability (3.9) we can express the simultaneous probability as

$$\begin{aligned} S(p_1, \dots, p_s, n_1, \dots, n_s) dp_1 \dots dp_s \\ = P(p_1, \dots, p_s) dp_1 \dots dp_s G(p_1, \dots, p_s; n_1, \dots, n_s) \\ = Q(n_1, \dots, n_s) H(n_1, \dots, n_s; p_1, \dots, p_s) dp_1 \dots dp_s, \end{aligned} \quad (3.10)$$

where $H(n_1, \dots, n_s; p_1, \dots, p_s) dp_1 \dots dp_s$ is the posterior probability and $Q(n_1, \dots, n_s)$ the marginal probability of observing the set of counts (n_1, \dots, n_s) independently of the values of the p 's. By integrating over the whole hyperspace of p 's we find

$$Q(n_1, \dots, n_s) \quad (3.11)$$

$$= \int_{p_1} \dots \int_{p_s} P(p_1, \dots, p_s) G(p_1, \dots, p_s; n_1, \dots, n_s) dp_1 \dots dp_s$$

since we must demand the normalization

$$\int_{p_1} \dots \int_{p_s} H(n_1, \dots, n_s; p_1, \dots, p_s) dp_1 \dots dp_s = 1 \quad (3.12)$$

Using (3.11) in (3.10), we finally obtain

$$\begin{aligned} & H(n_1, \dots, n_s; p_1, \dots, p_s) dp_1 \dots dp_s \\ &= \frac{P(p_1, \dots, p_s) G(p_1, \dots, p_s; n_1, \dots, n_s) dp_1 \dots dp_s}{\int_{p_1} \dots \int_{p_s} P(p_1, \dots, p_s) G(p_1, \dots, p_s; n_1, \dots, n_s) dp_1 \dots dp_s} \end{aligned} \quad (3.13)$$

If we have no information relating the p 's to each other, we must accept them as independent, which means that their joint distribution (3.9) can be written

$$P(p_1, \dots, p_r) dp_1 \dots dp_r = P(p_1) dp_1 \dots P(p_r) dp_r \quad (3.14)$$

Introducing this and (3.2) in formula (3.13), we find

$$\begin{aligned} & H(n_1, \dots, n_s; p_1, \dots, p_s) dp_1 \dots dp_s \\ &= \frac{P(p_1) G(p_1; n_1) dp_1}{\int_{p_1} P(p_1) G(p_1; n_1) dp_1} \dots \frac{P(p_s) G(p_s; n_s) dp_s}{\int_{p_s} P(p_s) G(p_s; n_s) dp_s} \\ &= H(n_1; p_1) dp_1 \dots H(n_s; p_s) dp_s \end{aligned} \quad (3.15)$$

Thus the posterior probability is in this case the product of the individual posterior probabilities.

For another example let us assume that we know that any p_i can be expressed as

$$p_i = q_i a \quad (3.16)$$

in such a way that a is independent of the value of any q_j . The limitations of the value of any q_j are

$$0 \leq q_j \leq 1 \quad (3.17)$$

and

$$q_1 + \dots + q_s = 1. \quad (3.18)$$

Since we are approaching the problem where a represents the same strength of a radioactive source, we will assume a constant a priori probability for a from zero to a high value K - which may tend to infinity (ref. III) - and thus

$$P(a)da = \begin{cases} 0 & \text{when } a > K \\ K^{-1} da & \text{" } 0 \leq a \leq K \\ 0 & \text{" } a < 0 \end{cases} \quad (3.19)$$

The information we have on the q 's is, besides the connection (3.18), that they are distribution probabilities and thus linearly additive. We therefore assume equal a priori probabilities for equal volumes in the space Ω defined by

$$q_1 + \dots + q_s \leq 1 \quad \text{and} \quad q_i \geq 0 \quad (3.20)$$

and obtain

$$P(q_1, \dots, q_s) dq_1 \dots dq_s = \times dq_1 \dots dq_{s-1} \delta(q_s - (1 - q_1 - \dots - q_{s-1})) dq_s \quad (3.21)$$

when the point (q_1, \dots, q_s) is in the space Ω

and

$$P(q_1, \dots, q_s) dq_1 \dots dq_s = 0 \quad (3.22)$$

when the point (q_1, \dots, q_s) is outside Ω . κ is a normalizing constant.

According to the assumed independency between a and the q 's we have

$$P(p_1, \dots, p_s) = P(a) da P(q_1, \dots, q_s) dq_1 \dots dq_s. \quad (3.23)$$

Introducing what we have found until now (equations (3.3), (3.19), (3.21), and (3.22)) in formulae (3.13), we find

$$H(n_1, \dots, n_s; a, q_1, \dots, q_s) da dq_1 \dots dq_s \quad (3.24)$$

$$= \frac{\delta(q_s - (1 - q_1 - \dots - q_{s-1})) \frac{a^n}{n!} e^{-a} \frac{n!}{n_1! \dots n_s!} q_1^{n_1} \dots q_s^{n_s} da dq_1 \dots dq_s}{\int_0^\infty \frac{a^n}{n!} e^{-a} da \int \dots \int_\Omega \delta(q_s - (1 - q_1 - \dots - q_{s-1})) \frac{n!}{n_1! \dots n_s!} q_1^{n_1} \dots q_s^{n_s} dq_1 \dots dq_s}.$$

Here

$$\int_0^\infty \frac{a^n}{n!} e^{-a} da = 1 \quad (3.25)$$

and

$$\begin{aligned} & \int \dots \int_\Omega \delta(q_s - (1 - q_1 - \dots - q_{s-1})) q_1^{n_1} \dots q_s^{n_s} dq_1 \dots dq_s \\ &= \int \dots \int_{\Omega^*} dq_1 \dots dq_{s-2} \int_{q_{s-1}=0}^{1-(q_1+\dots+q_{s-2})} q_{s-1}^{n_{s-1}} (q_1+\dots+q_{s-2})^{n_s} dq_{s-1} \\ & \quad (3.26) \end{aligned}$$

$$= \int \dots \int_{\Omega^*} dq_1 \dots dq_{s-2} (1 - (q_1 + \dots + q_{s-2}))^{n_s + n_{s-1} + 1} \int_{u=0}^1 u^{n_{s-1}-1} (1-u)^{n_s} du,$$

which by repeated use of substitution of the kind

$$q_{s-1} = u(1-(q_1+\dots+q_{s-2})) \quad (3.27)$$

and the integral

$$\int_{u=0}^1 u^{n_1}(1-u)^{n_2} du = \frac{n_1! n_2!}{(n_1+n_2+1)!} \quad (3.28)$$

is reduced to

$$\begin{aligned} & \int_{\Omega} \dots \int \delta(q_s - (1-q_1-\dots-q_{s-1})) q_1^{n_1} \dots q_s^{n_s} dq_1 \dots dq_s \\ &= \frac{n_1! \dots n_s!}{(n_1+\dots+n_s+s-1)!} \end{aligned} \quad (3.29)$$

We therefore obtain as the final result

$$\begin{aligned} & H(n_1, \dots, n_s; a, q_1, \dots, q_s) da dq_1 \dots dq_s \\ &= \delta(q_s - (1-q_1-\dots-q_{s-1})) \frac{a^n}{n!} e^{-a} \frac{(n+s-1)!}{n_1! \dots n_s!} q_1^{n_1} \dots q_s^{n_s} da dq_1 \dots dq_s \end{aligned} \quad (3.30)$$

if $a \geq 0$ and (q_1, \dots, q_s) is inside Ω , and

$$H(n_1, \dots, n_s; a, q_1, \dots, q_s) da dq_1 \dots dq_s = 0 \quad (3.31)$$

if $a < 0$ and/or (q_1, \dots, q_s) is outside Ω .

It is now possible to answer the question of the mean value of q_1 .

We find

$$E[q_1] = \frac{(n+s-1)!}{n_1! \dots n_s!} \cdot \frac{n_1! \dots (n_1+1)! \dots n_s!}{(n+s-1+1)!} = \frac{n_1+1}{n+s} \quad (3.32)$$

The square of the standard deviation $D[q_1]$ can be found as follows:

$$\begin{aligned}
D^2 \{ q_i \} &= E \{ q_i^2 \} - E^2 \{ q_i \} \\
&= \frac{(n_i+1)(n_i+2)}{(n+s)(n+s+1)} - \frac{(n_i+1)^2}{(n+s)^2} \\
&= \frac{(n+s)(n_i+1) - (n_i+1)^2}{(n+s)^2 (n+s+1)} \rightarrow 0
\end{aligned} \tag{3.33}$$

as the total count number $n \rightarrow \infty$, since the numerator is of the second power in n and the denominator of the third power.

This shows that the mean value $E[q_i]$ given by (3.32) in the limit of very great total number of observations tends to give exact information on the value of q_i .

In the other limit, where no observations have been performed, we see that the mean value of q_i is a constant

$$E \{ q_i \} = \frac{1}{s} \tag{3.34}$$

and this is identical with the original postulate (Bayes 1763) on equal a priori probabilities of all possible kinds of events.

By putting the total count number n equal to zero in formula (3.29) we also see that the normalizing constant κ in (3.21) must be

$$\kappa = (s-1)! \tag{3.35}$$

CHAPTER 4:

THE β - γ -COINCIDENCE METHOD

In the type of instrument we shall investigate in this chapter the decays in the radioactive source A may be detected by means of two detectors of different kinds. We shall denote them the β - and the γ -detector.

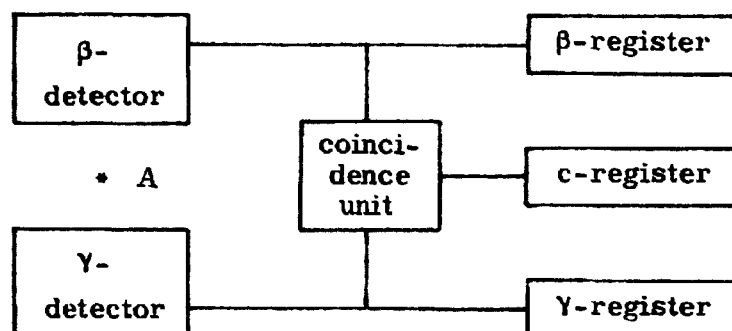


Fig. 4.1. Detectors Electronics and scalers

Figure 4.1 shows schematically how the two detectors are connected to the three registers. A single decay of a radioactive nucleus in the source A may here cause one of eight possible types of events. By an event we mean a combination of registrations in the three scales shown.

Event no.	1	2	3	4	5	6	7	8
β-register	+	+	+	+	0	0	0	0
Y-register	+	+	0	0	+	+	0	0
c-register	+	0	+	0	+	0	+	0

Fig. 4.2

Figure 4.2 shows all these eight possible events, and we must now examine them one by one. The idea of the c-register is that it should accumulate the number of events for which - for the same decay - registrations have been made in the two other scalars. This is true of event no. 1, but in event no. 2 there have been registrations in the two other scalars, but not in the coincidence scalars, and in events nos. 3, 5, and 7 we see registrations in this register without simultaneous registrations in the β- and Y-scalars. In designing the coincidence circuit we must therefore exclude the possibility of events nos. 2, 3, 5, and 7.

In order to exclude event no. 2 we must have a resolving time in the coincidence circuit long enough to ensure that all decays giving β- and Y-registrations are registered in the c-register even if one or both of the detectors should exhibit time fluctuations. A systematical time delay in one

of the detectors is offset by an artificial delay circuit in the other detector.

It is in general easier to ensure that a registration in the coincidence register produces registrations in both of the two scalers, and this excludes events nos. 3, 5 and 7.

The long resolving time that had to be introduced at this point will naturally cause accidental coincidence where a β -registration from one decay occurs within the resolving time simultaneously with a γ -registration from another decay. In chapter 2, however, we found that for other reasons it was necessary to assume very low counting rates; thus we just have to point out that here we have one reason more.

If all these demands are fulfilled, we have only four possible types of events following each decay; these types are shown in fig. 4.3.

Event no.	1	2	3	4
β -register	+	+	0	0
γ -register	+	0	+	0
c-register	+	0	0	0

Fig. 4.3

Let us denote by n_i the accumulated number of registrations during an observation period of event no. i . Then the number n_β of registrations in the β -scaler is

$$n_\beta = n_1 + n_2, \quad (4.1)$$

and corresponding to this

$$n_\gamma = n_1 + n_3 \quad (4.2)$$

and

$$n_c = n_1. \quad (4.3)$$

From this we obtain

$$\begin{aligned} n_1 &= n_c \\ n_2 &= n_\beta - n_c \\ n_3 &= n_\gamma - n_c. \end{aligned} \quad (4.4)$$

We have no observation of n_4 .

We may now have the further information on the nature of the decay and the design of the detectors that the probability that a decay is detected by one of the detectors and registered in the corresponding register is independent of whether the same decay causes a registration in the other register or not. This means that the two detection and registration probabilities are independent, and thus the probability of a simultaneous registration is the mere product of these two probabilities.

With the notation already introduced by (2.2) we have the detection and registration probabilities in the β - and γ -register respectively:

$$\varepsilon_{\beta}(x, y, z, t) \quad (4.5)$$

and

$$\varepsilon_{\gamma}(x, y, z, t) . \quad (4.6)$$

For the probability, q_1 , of having the event of a coincidence we can write, assuming the above-mentioned information,

$$q_1 = \varepsilon_c(x, y, z, t) = \varepsilon_{\beta}(x, y, z, t) \cdot \varepsilon_{\gamma}(x, y, z, t) . \quad (4.7)$$

The probability, q_2 , of obtaining a β -registration without a simultaneous γ -registration (event no. 2 in fig. 4.3) is then

$$q_2 = \varepsilon_{\beta}(x, y, z, t) \{ 1 - \varepsilon_{\gamma}(x, y, z, t) \} , \quad (4.8)$$

and similarly for event no. 3:

$$q_3 = \varepsilon_{\gamma}(x, y, z, t) \{ 1 - \varepsilon_{\beta}(x, y, z, t) \} . \quad (4.9)$$

Actual radioactive sources are usually not point sources with constant activity, but it is in general possible to assume that the distribution of the activity may be written as

$$A(x, y, z, t) = A_1(x, y, z) \cdot A_2(t) . \quad (4.10)$$

This means that the radioactivity is distributed in space, but that all points of the thus extended source exhibit the same variation in time (e.g. the half-life is the same in all parts of the source).

The simple relation (4.7) for each point of the source will not in general remain unchanged under the integration over volume and time which gave us (2.21). But if we further assume that the registration probability is independent of time in one of the detectors (e. g. the β -detector) and independent of space in one (e. g. the γ -detector), we have

$$\epsilon_{\beta}(x, y, z, t) = \epsilon_{\beta}(x, y, z) \quad (4.11)$$

and

$$\epsilon_{\gamma}(x, y, z, t) = \epsilon_{\gamma}(t) \quad (4.12)$$

If we use (4.10), (4.11) and (4.12) in (2.21), we find

$$\bar{\epsilon}_{\beta} = \frac{\iiint_{\Omega} \epsilon_{\beta}(x, y, z) A_1(x, y, z) dx dy dz \int_0^{t'} A_2(t) dt}{\iiint_{\Omega} A_1(x, y, z) dx dy dz \int_0^{t'} A_2(t) dt} \quad (4.13)$$

$$\bar{\epsilon}_{\gamma} = \frac{\iiint_{\Omega} A_1(x, y, z) dx dy dz \int_0^{t'} \epsilon_{\gamma}(t) A_2(t) dt}{\iiint_{\Omega} A_1(x, y, z) dx dy dz \int_0^{t'} A_2(t) dt} \quad (4.14)$$

and

$$\begin{aligned} \bar{\epsilon}_c &= \frac{\iiint_{\Omega} \epsilon_{\beta}(x, y, z) A_1(x, y, z) dx dy dz \int_0^{t'} \epsilon_{\gamma}(t) A_2(t) dt}{\iiint_{\Omega} A_1(x, y, z) dx dy dz \int_0^{t'} A_2(t) dt} \\ &= \bar{\epsilon}_{\beta} \cdot \bar{\epsilon}_{\gamma} \end{aligned} \quad (4.15)$$

We thus see that only one of the detectors need be stable in time and only one need have a constant detection probability over the whole extension of the radioactive source in order to maintain the relation (4.7).

This result was reported by J. Putman (1950), but later denied by G. Wolf (1960).

In the present case we have a slightly different problem, since neither the β - nor the γ -detector exhibits a constant detection probability over the whole extension of the radioactive source. But the gold foil we are going to measure was irradiated perpendicularly in a well collimated beam of thermal neutrons freed from epithermal neutrons by means of a bismuth filter. If we therefore introduce a co-ordinate system like that in figure 4.4, in which we can describe the activity distribution in the gold foil, we see

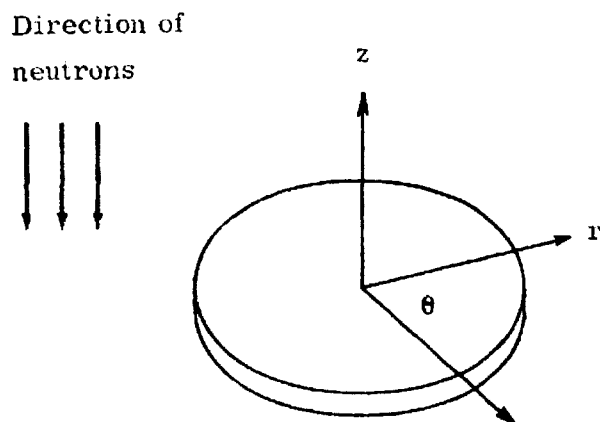


Fig. 4.4. Geometry of a gold foil.

that the activity at a point (r, θ, z) at the time t can be written as

$$A(r, \theta, z, t) = A_1(r, \theta) A_2(z) A_3(t) , \quad (4.16)$$

where $A_1(r, \theta)$ describes the inhomogeneity of the beam, $A_2(z)$ describes the attenuation of the beam passing the foil, and $A_3(t)$ in the exponential function describes the decay of the radioactive source strength.

Now it is shown in the description of the experimental arrangement that the gamma detector has a constant detection probability in the z direction, and that the beta detector is insensitive to movements in the (r, θ) plane. Both detectors are very stable in time, but it is enough to state that the beta detector is stable. We have

$$\epsilon_{\gamma}(r, \theta, z, t) = \epsilon_{\gamma}(r, \theta, t) \quad (4.17)$$

and

$$\epsilon_{\beta}(r, \theta, z, t) = \epsilon_{\beta}(z) . \quad (4.18)$$

Introducing (4.16), (4.17) and (4.13) in (2.21), we find

$$\bar{\epsilon}_\beta = \frac{\int_z \epsilon_\beta(z) A_2(z) dz \int_r \int_\theta \int_t A_1(r, \theta) A_3(t) r dr d\theta dt}{\int_z A_2(z) dz \int_r \int_\theta \int_t A_1(r, \theta) A_3(t) r dr d\theta dt} \quad (4.19)$$

$$\bar{\epsilon}_\gamma = \frac{\int_r \int_\theta \int_t \epsilon_\gamma(r, \theta, t) A_1(r, \theta) A_3(t) r dr d\theta dt \int_z A_2(z) dz}{\int_r \int_\theta \int_t A_1(r, \theta) A_3(t) r dr d\theta dt \int_z A_2(z) dz} \quad (4.20)$$

$$\begin{aligned} \text{and} \quad \bar{\epsilon}_c &= \frac{\int_z \epsilon_\beta(z) A_2(z) dz \int_r \int_\theta \int_t \epsilon_\gamma(r, \theta, t) A_1(r, \theta) A_3(t) r dr d\theta dt}{\int_z A_2(z) dz \int_r \int_\theta \int_t A_1(r, \theta) A_3(t) r dr d\theta dt} \\ &= \bar{\epsilon}_\beta \cdot \bar{\epsilon}_\gamma. \end{aligned} \quad (4.21)$$

Thus we see that also under these conditions our simple relation (4.7) is maintained.

We are therefore able to reformulate our problem in a simpler formalism since we do not have to bother more about (x, y, z) and t .

The parameters in question are the strength or absolute activity a of the radioactive source as given by (2.20), and the two detection and registration probabilities ϵ_β and ϵ_γ , which are both mean values over space and time. We have the further knowledge that the coincidence probability ϵ_c is the product of ϵ_β and ϵ_γ (4.21). Our observed quantities are the numbers of registrations, n_β , n_γ and n_c , in the respective scalers, and the number n_i of events of the i^{th} type (fig. 4.3) is given by (4.4)

Hence the probability distribution of the observed count numbers, as given by (2.17) and with the use of the a priori information (4.21), will be

$$G(a, \varepsilon_\beta, \varepsilon_\gamma; n_1, n_2, n_3) \quad (4.22)$$

$$= \frac{\{a \varepsilon_\beta \varepsilon_\gamma\}^{n_1}}{n_1!} e^{-a \varepsilon_\beta \varepsilon_\gamma} \frac{\{a \varepsilon_\beta (1 - \varepsilon_\gamma)\}^{n_2}}{n_2!} e^{-a \varepsilon_\beta (1 - \varepsilon_\gamma)} \frac{\{a \varepsilon_\gamma (1 - \varepsilon_\beta)\}^{n_3}}{n_3!} e^{-a \varepsilon_\gamma (1 - \varepsilon_\beta)}$$

independently of the value of n_4 .

The activity can have any positive value smaller than a big number K ; therefore the a priori probability is again assumed to be

$$P(a) da = \frac{da}{K} \quad \text{when } 0 \leq a < K \quad (4.23)$$

and $P(a) da = 0$ otherwise.

K may tend to infinity, and this will be written in the following without further comment.

The two probabilities ε_β and ε_γ may, independently of each other, be given any value between zero and unity; so we have

$$P(\varepsilon_\beta) d\varepsilon_\beta = d\varepsilon_\beta \quad \text{when } 0 \leq \varepsilon_\beta \leq 1 \quad (4.24)$$

and $P(\varepsilon_\beta) d\varepsilon_\beta = 0$ otherwise,

and similarly

$$P(\varepsilon_\gamma) d\varepsilon_\gamma = d\varepsilon_\gamma \quad \text{when } 0 \leq \varepsilon_\gamma \leq 1 \quad (4.25)$$

and $P(\varepsilon_\gamma) d\varepsilon_\gamma = 0$ otherwise.

Now we have all we need to derive the posterior probability, and using (4.22), (4.23), (4.24), and (4.25) in (3.13), we find

$$\begin{aligned} & H(n_1, n_2, n_3; a, \varepsilon_\beta, \varepsilon_\gamma) da d\varepsilon_\beta d\varepsilon_\gamma \\ &= \frac{G(a, \varepsilon_\beta, \varepsilon_\gamma; n_1, n_2, n_3) da d\varepsilon_\beta d\varepsilon_\gamma}{\int_{a=0}^{\infty} \int_{\varepsilon_\beta=0}^1 \int_{\varepsilon_\gamma=0}^1 G(a, \varepsilon_\beta, \varepsilon_\gamma; n_1, n_2, n_3) da d\varepsilon_\beta d\varepsilon_\gamma} \end{aligned} \quad (4.26)$$

when

$$a \geq 0, \quad 0 \leq \epsilon_\beta \leq 1 \quad \text{and} \quad 0 \leq \epsilon_\gamma \leq 1,$$

and

$$H(n_1, n_2, n_3; a, \epsilon_\beta, \epsilon_\gamma) da d\epsilon_\beta d\epsilon_\gamma = 0 \quad \text{otherwise.}$$

Evaluation of the Integral in the Denominator

The integral I in the denominator of (4.26) is given by

$$\begin{aligned} I &= \int_{a=0}^{\infty} \int_{\epsilon_\beta=0}^1 \int_{\epsilon_\gamma=0}^1 G(a, \epsilon_\beta, \epsilon_\gamma; n_1, n_2, n_3) da d\epsilon_\beta d\epsilon_\gamma \\ &= \iiint_a^{n_1+n_2+n_3} e^{-a \{ \epsilon_\beta \epsilon_\gamma + \epsilon_\beta(1-\epsilon_\gamma) + \epsilon_\gamma(1-\epsilon_\beta) \}} \frac{1}{n_1! n_2! n_3!} \\ &\quad (\epsilon_\beta \epsilon_\gamma)^{n_1} (\epsilon_\beta(1-\epsilon_\gamma))^{n_2} (\epsilon_\gamma(1-\epsilon_\beta))^{n_3} da d\epsilon_\beta d\epsilon_\gamma \\ &= \frac{1}{n_1! n_2! n_3!} \iiint_a^{n_1+n_2+n_3} e^{-a \{ 1 - (1-\epsilon_\beta)(1-\epsilon_\gamma) \}} \\ &\quad \epsilon_\beta^{n_1+n_2} (1-\epsilon_\beta)^{n_3} \epsilon_\gamma^{n_1+n_3} (1-\epsilon_\gamma)^{n_2} da d\epsilon_\beta d\epsilon_\gamma. \end{aligned} \tag{4.27}$$

At this place we use the substitutions

$$\begin{aligned} x &= 1 - \epsilon_\beta & dx &= -d\epsilon_\beta & 0 \leq \epsilon_\beta \leq 1 & \text{ gives } 1 \geq x \geq 0 \\ y &= 1 - \epsilon_\gamma & dy &= -d\epsilon_\gamma & 0 \leq \epsilon_\gamma \leq 1 & \text{ gives } 1 \geq y \geq 0. \end{aligned}$$

Thus we get

$$I = \frac{1}{n_1! n_2! n_3!} \int_{a=0}^{\infty} \int_{x=0}^1 \int_{y=0}^1 a^{n_1+n_2+n_3} e^{-a} e^{axy} (1-x)^{n_1+n_2} x^{n_3} (1-y)^{n_1+n_3} y^{n_2} da dx dy . \quad (4.28)$$

In (4.28) we now expand the mixed term by a power series

$$e^{axy} = \sum_{k=0}^{\infty} \frac{(axy)^k}{k!} \quad (4.29)$$

so that a separation of the variables will be possible, and we find, using the fact that the order of summation and integration can be interchanged,

$$\begin{aligned} I &= \frac{1}{n_1! n_2! n_3!} \sum_{k=0}^{\infty} \frac{1}{k!} \int_0^{\infty} a^{n_1+n_2+n_3+k} e^{-a} da \int_0^1 x^{n_3+k} (1-x)^{n_1+n_2} dx \\ &\quad \int_0^1 y^{n_2+k} (1-y)^{n_1+n_3} dy \\ &= \frac{1}{n_1! n_2! n_3!} \sum_{k=0}^{\infty} \frac{1}{k!} \Gamma(n_1+n_2+n_3+k+1) \frac{\Gamma(n_3+k+1) \Gamma(n_1+n_2+1)}{\Gamma(n_1+n_2+n_3+k+2)} \\ &\quad \frac{\Gamma(n_2+k+1) \Gamma(n_1+n_3+1)}{\Gamma(n_1+n_2+n_3+k+2)} \\ &= \frac{\Gamma(n_1+n_2+1) \Gamma(n_1+n_3+1)}{n_1! n_2! n_3!} \sum_{k=0}^{\infty} \frac{\Gamma(n_2+k+1) \Gamma(n_3+k+1)}{\Gamma(n_1+n_2+n_3+k+2) k!} \frac{1}{(n_1+n_2+n_3+k+1)} . \end{aligned} \quad (4.30)$$

Here we have used the formulae (e. g. Standard Mathematical Tables, 1954)

$$\int_0^{\infty} x^n e^{-x} dx = \Gamma(n+1) \quad (4.31)$$

and

$$\int_0^1 x^m (1-x)^n dx = \frac{\Gamma(m+1) \Gamma(n+1)}{\Gamma(m+n+2)} . \quad (4.32)$$

As the value of the hypergeometric series in which $c - b - a > 0$ is

$$\begin{aligned} F(a, b; c; 1) &= \frac{\Gamma(c)}{\Gamma(a) \Gamma(b)} \sum_{k=0}^{\infty} \frac{\Gamma(a+k) \Gamma(b+k)}{\Gamma(c+k) k!} \\ &= \frac{\Gamma(c) \Gamma(c-b-a)}{\Gamma(c-a) \Gamma(c-b)} \end{aligned} \quad (4.33)$$

(Whittaker and Watson, 1963), we find

$$\sum_{k=0}^{\infty} \frac{\Gamma(a+k) \Gamma(b+k)}{\Gamma(c+k) k!} = \frac{\Gamma(a) \Gamma(b) \Gamma(c-b-a)}{\Gamma(c-a) \Gamma(c-b)} . \quad (4.34)$$

Thus we see that the only difficulty in evaluating the series (4.30) is the factor 1 divided by $n_1 + n_2 + n_3 + k + 1$. We therefore expand this term in the following way:

$$\begin{aligned} \frac{1}{n_1 + n_2 + n_3 + k + 1} &= \frac{1}{n_1 + n_2 + n_3 + k + 2} \left\{ 1 + \frac{1}{n_1 + n_2 + n_3 + k + 1} \right\} \\ &= \frac{1}{n_1 + n_2 + n_3 + k + 2} + \frac{1}{(n_1 + n_2 + n_3 + k + 2)(n_1 + n_2 + n_3 + k + 3)} \left\{ 1 + \frac{1}{n_1 + n_2 + n_3 + k + 1} \right\} \\ &= \sum_{i=0}^{\infty} \frac{i!}{(n_1 + n_2 + n_3 + 2 + k) \dots (n_1 + n_2 + n_3 + 2 + i + k)} . \end{aligned} \quad (4.35)$$

Insertion of this in (4.30) gives

$$\begin{aligned}
 I &= \frac{\Gamma(n_1+n_2+1) \Gamma(n_1+n_3+1)}{n_1! n_2! n_3!} \sum_{i=0}^{\infty} i! \sum_{k=0}^{\infty} \frac{\Gamma(n_2+k+1) \Gamma(n_3+k+1)}{\Gamma(n_1+n_2+n_3+k+2+i) k!} \\
 &= \frac{\Gamma(n_1+n_2+1) \Gamma(n_1+n_3+1)}{n_1! n_2! n_3!} \sum_{i=0}^{\infty} i! \frac{\Gamma(n_2+1) \Gamma(n_3+1) \Gamma(n_1+i)}{\Gamma(n_1+n_3+1+i) \Gamma(n_1+n_2+1+i)} \\
 &= \frac{\Gamma(n_1+n_2+1) \Gamma(n_1+n_3+1)}{n_1!} \sum_{i=0}^{\infty} \frac{\Gamma(1+i) \Gamma(n_1+i)}{\Gamma(n_1+n_3+1+i) \Gamma(n_1+n_2+1+i)} .
 \end{aligned} \tag{4.36}$$

This is also a generalized hypergeometric series, but one converging much more rapidly, the first terms being

$$\begin{aligned}
 I &= \frac{1}{(n_1+n_2+1)(n_1+n_3+1)} \\
 &+ \frac{n_1+1}{(n_1+n_2+1)(n_1+n_2+2)(n_1+n_3+1)(n_1+n_3+2)} + o\left(\frac{1}{n_1^4}\right) ,
 \end{aligned} \tag{4.37}$$

where $o\left(\frac{1}{n_1^4}\right)$ is a function that goes to zero at least as rapidly as 1 divided by n_1^4 when n_1 goes to infinity.

We have thus found the posterior probability function (4.26) to be expressed by

$$\begin{aligned}
 H(n_1, n_2, n_3; a, \epsilon_\beta, \epsilon_\gamma) da d\epsilon_\beta d\epsilon_\gamma \\
 = \frac{1}{\Gamma} G(a, \epsilon_\beta, \epsilon_\gamma; n_1, n_2, n_3) da d\epsilon_\beta d\epsilon_\gamma
 \end{aligned} \tag{4.38}$$

when $a \geq 0$, $0 \leq \epsilon_\beta \leq 1$ and $0 \leq \epsilon_\gamma \leq 1$,

and $H(n_1, n_2, n_3; a, \epsilon_\beta, \epsilon_\gamma) da d\epsilon_\beta d\epsilon_\gamma = 0$ otherwise.

$G(a, \epsilon_\beta, \epsilon_\gamma; n_1, n_2, n_3)$ is given by (4.22) and I by (4.36).

Some Figures Characterizing the Function (4.38)

The function (4.38), which gives the simultaneous probability distribution of the unknown parameters a , ϵ_β and ϵ_γ , can be described in many ways, and some of them will be shown here. In fact the function (4.38) itself contains all the information we have on the value of the parameters when we have observed the count numbers n_1 , n_2 and n_3 .

Nevertheless we will here calculate a few figures that may describe the function (4.38); we begin with the mean value of a ,

$$\begin{aligned}
 E[a] &= \int_{a=0}^{\infty} \int_{\epsilon_\beta=0}^1 \int_{\epsilon_\gamma=0}^1 a H(n_1, n_2, n_3; a, \epsilon_\beta, \epsilon_\gamma) da d\epsilon_\beta d\epsilon_\gamma \\
 &= \frac{1}{\Gamma(n_1)! \Gamma(n_2)! \Gamma(n_3)!} \sum_{k=0}^{\infty} \frac{1}{k!} \int_0^{\infty} a^{n_1+n_2+n_3+1+k} e^{-a} da \int_0^1 x^{n_3+k} (1-x)^{n_1+n_2} dx \\
 &\quad \int_0^1 y^{n_2+k} (1-y)^{n_1+n_3} dy \\
 &= \frac{1}{\Gamma(n_1)! \Gamma(n_2)! \Gamma(n_3)!} \sum_{k=0}^{\infty} \frac{1}{k!} \Gamma(n_1+n_2+n_3+2+k) \frac{\Gamma(n_3+k+1) \Gamma(n_1+n_2+1)}{\Gamma(n_1+n_2+n_3+2+k)} \\
 &\quad \frac{\Gamma(n_2+k+1) \Gamma(n_1+n_3+1)}{\Gamma(n_1+n_2+n_3+2+k)} \\
 &= \frac{\Gamma(n_1+n_2+1) \Gamma(n_1+n_3+1)}{\Gamma(n_1)! \Gamma(n_2)! \Gamma(n_3)!} \times \frac{\Gamma(n_3+1) \Gamma(n_2+1) \Gamma(n_1)}{\Gamma(n_1+n_2+1) \Gamma(n_1+n_3+1)} \\
 &= \frac{1}{\Gamma(n_1)} .
 \end{aligned} \tag{4.39}$$

If $n_1 = 0$, we have a case ($c-a-b = 0$) where the hypergeometric series does not converge, and in this case the mean value of a is not defined.

By using the relations (4.37), (4.1), (4.2), and (4.3) in (4.39) we find

$$E[a] \approx \frac{(n_1+n_2+1)(n_1+n_3+1)}{n_1} \approx \frac{n_\beta}{n_c} \frac{n_\gamma}{n_c}, \quad (4.40)$$

which is the familiar formula. The error in the approximations is only 1 divided by n_c and is negligible when compared with the relative standard deviation, which is of the order of 1 divided by the square root of n_c .

In order to find the standard deviation of a we first calculate

$$\begin{aligned} E[a^2] &= \int_{a=0}^{\infty} \int_{\beta=0}^1 \int_{\gamma=0}^1 a^2 H(n_1, n_2, n_3; a, \beta, \gamma) da d\beta d\gamma \\ &= \frac{1}{n_1! n_2! n_3!} \sum_{k=0}^{\infty} \frac{1}{k!} \int_0^{\infty} a^{n_1+n_2+n_3+2+k} e^{-a} da \int_0^1 x^{n_3+k} (1-x)^{n_1+n_2} dx \\ &\quad \int_0^1 y^{n_2+k} (1-y)^{n_1+n_3} dy \\ &= \frac{1}{n_1! n_2! n_3!} \sum_{k=0}^{\infty} \frac{1}{k!} \Gamma(n_1+n_2+n_3+3+k) \frac{\Gamma(n_2+1+k) \Gamma(n_1+n_3+1)}{\Gamma(n_1+n_2+n_3+2+k)} \\ &\quad \frac{\Gamma(n_3+1+k) \Gamma(n_1+n_2+1)}{\Gamma(n_1+n_2+n_3+2+k)} \\ &= \frac{\Gamma(n_1+n_2+1) \Gamma(n_1+n_3+1)}{n_1! n_2! n_3!} \sum_{k=0}^{\infty} \frac{\Gamma(n_3+1+k) \Gamma(n_2+1+k)}{\Gamma(n_1+n_2+n_3+2+k) k!} (n_1+n_2+n_3+2+k) \\ &= \frac{\Gamma(n_1+n_2+1) \Gamma(n_1+n_3+1)}{n_1! n_2! n_3!} \left\{ \sum_{k=0}^{\infty} \frac{\Gamma(n_3+1+k) \Gamma(n_2+1+k)}{\Gamma(n_1+n_2+n_3+2+k) k!} + \sum_{k=0}^{\infty} \frac{\Gamma(n_3+1+k) \Gamma(n_2+1+k)}{\Gamma(n_1+n_2+n_3+2+k) k!} \right\} \end{aligned}$$

$$\begin{aligned}
&= \frac{\Gamma(n_1+n_2+1) \Gamma(n_1+n_3+1)}{\Gamma n_1! \Gamma n_2! \Gamma n_3!} \left\{ \frac{\Gamma(n_3+1) \Gamma(n_2+1) \Gamma(n_1-1)}{\Gamma(n_1+n_2) \Gamma(n_1+n_3)} + \frac{\Gamma(n_3+1) \Gamma(n_2+1) \Gamma(n_1)}{\Gamma(n_1+n_2+1) \Gamma(n_1+n_3+1)} \right\} \\
&= \frac{1}{\Gamma} \left\{ \frac{(n_1+n_2)(n_1+n_3)}{n_1(n_1-1)} + \frac{1}{n_1} \right\}. \quad (4.41)
\end{aligned}$$

From (4.41) and (4.39) we now find the squared standard deviation by

$$\begin{aligned}
D^2[a] &= E[a^2] - E^2[a] \\
&= \frac{1}{\Gamma} \left\{ \frac{(n_1+n_2)(n_1+n_3)}{n_1(n_1-1)} + \frac{1}{n_1} \right\} - \frac{1}{\Gamma^2 n_1^2} \\
&= E^2[a] \left\{ \Gamma \frac{n_1(n_1+n_2)(n_1+n_3)}{n_1-1} + \Gamma n_1 - 1 \right\}. \quad (4.42)
\end{aligned}$$

Using (4.37) again, we find the square of the relative standard deviation to be

$$\begin{aligned}
\frac{D^2[a]}{E^2[a]} &= \frac{n_1(n_1+n_2)(n_1+n_3)}{(n_1+n_2+1)(n_1+n_3+1)(n_1-1)} \\
&\quad + \frac{n_1(n_1+1)(n_1+n_2)(n_1+n_3)}{(n_1+n_2+1)(n_1+n_3+1)(n_1+n_2+2)(n_1+n_3+2)(n_1-1)} \\
&\quad + \frac{1}{(n_1+n_2+1)(n_1+n_3+1)} - 1 + \sigma\left(\frac{1}{n_1^2}\right), \quad (4.43)
\end{aligned}$$

where we have collected terms small to 1 divided by n_1^2 . By some tedious and uninteresting calculations (4.43) can be written as

$$\frac{D^2[a]}{E^2[a]} = \frac{1}{n_1} \left\{ \frac{2 n_1^2}{(n_1+n_2+1)(n_1+n_3+1)} - \frac{n_1}{(n_1+n_3+1)} - \frac{n_1}{(n_1+n_2+1)} + 1 + \sigma\left(\frac{1}{n_1}\right) \right\}$$

$$\approx \frac{1}{n_c} \left\{ \frac{2 n_c^2}{n_\beta n_\gamma} - \frac{n_c}{n_\gamma} - \frac{n_c}{n_\beta} + 1 \right\}, \quad (4.44)$$

which is very close to the formula given by Campion (1960):

$$\frac{D^2[a]}{E^2[a]} \approx \frac{1}{n_c} \left\{ 2 \varepsilon_\beta \varepsilon_\gamma - \varepsilon_\beta - \varepsilon_\gamma + 1 \right\}. \quad (4.45)$$

The difference between (4.44) and (4.45) is, however, that the former derived on the basis of this work, only includes observed quantities, whereas the latter contains the values of ε_β and ε_γ , and we did not know these values.

We can of course calculate the mean value, $E[\varepsilon_\beta]$, on the basis of (4.38), and we find

$$E[\varepsilon_\beta] = \int_{a=0}^{\infty} \int_{\varepsilon_\beta=0}^{\infty} \int_{\varepsilon_\gamma=0}^{\infty} \varepsilon_\beta H(n_1, n_2, n_3; a, \varepsilon_\beta, \varepsilon_\gamma) da d\varepsilon_\beta d\varepsilon_\gamma$$

$$= \frac{1}{1 n_1! n_2! n_3!} \sum_{k=0}^{\infty} \frac{1}{k!} \int_0^{\infty} a^{n_1+n_2+n_3+k} e^{-a} da \int_0^1 x^{n_3+k} (1-x)^{n_1+n_2+1} dx$$

$$\int_0^1 y^{n_2+k} (1-y)^{n_1+n_3} dy$$

$$= \frac{1}{1 n_1! n_2! n_3!} \sum_{k=0}^{\infty} \frac{1}{k!} \Gamma(n_1+n_2+n_3+k+1) \frac{\Gamma(n_3+k+1) \Gamma(n_1+n_2+2)}{\Gamma(n_1+n_2+n_3+k+3)}$$

$$\frac{\Gamma(n_2+k+1) \Gamma(n_1+n_3+1)}{\Gamma(n_1+n_2+n_3+k+2)}$$

$$= \frac{\Gamma(n_1+n_2+2) \Gamma(n_1+n_3+1)}{\Gamma n_1! \Gamma n_2! \Gamma n_3!} \sum_{k=0}^{\infty} \frac{\Gamma(n_3+k+1) \Gamma(n_2+k+1)}{\Gamma(n_1+n_2+n_3+k+3) k!} \frac{1}{(n_1+n_2+n_3+k+1)} .$$

(4.46)

As in the derivation of (4.35) we can now write

$$\begin{aligned} \frac{1}{n_1+n_2+n_3+k+1} &= \frac{1}{n_1+n_2+n_3+k+3} \left\{ 1 + \frac{2}{n_1+n_2+n_3+k+1} \right\} \\ &= \frac{1}{n_1+n_2+n_3+k+3} + o\left(\frac{1}{n_1}\right) , \end{aligned}$$

(4.47)

and introducing this in (4.46), we obtain

$$\begin{aligned} E[\epsilon_\beta] &= \frac{\Gamma(n_1+n_2+2) \Gamma(n_1+n_3+1)}{\Gamma n_1! \Gamma n_2! \Gamma n_3!} \frac{\Gamma(n_3+1) \Gamma(n_2+1) \Gamma(n_1+2)}{\Gamma(n_1+n_2+3) \Gamma(n_1+n_3+3)} + o\left(\frac{1}{n_1}\right) \\ &= \frac{n_1+1}{\Gamma(n_1+n_2+2) \Gamma(n_1+n_3+1) \Gamma(n_1+n_3+2)} + o\left(\frac{1}{n_1}\right) \\ &= \frac{n_1}{n_1+n_3} + o\left(\frac{1}{n_1}\right) \\ &\approx \frac{n_c}{n_Y} . \end{aligned}$$

(4.48)

In a similar way we find

$$E[\epsilon_Y] \approx \frac{n_c}{n_\beta} ,$$

(4.49)

and both formulae are well known.

CHAPTER 5:

DESCRIPTION OF THE ACTUAL EXPERIMENT

To demonstrate in detail that the absolute measurement of radioactive source strength made during the experiment to be described is fully covered by the theoretical evaluation given above, we must review the individual components forming the complete experimental set-up. These components are

- (1) the nature of the radioactive source and the special features of the radionuclide in question (here Au^{198});
- (2) the design of the two detectors;
- (3) the electronic circuit with emphasis on the coincidence part.

Finally the action of the whole system must be demonstrated.

The Decay of Au^{198}

In connection with the standardization of a neutron beam (Als-Nielsen, 1966) absolute measurement of Au^{198} was required, and although we have used the coincidence method for other radionuclides, we will here use Au^{198} as an example for demonstration. The decay scheme of Au^{198} is shown in figure 5.1 (Nuclear Data Sheet), and we must examine whether the assumption which gave the simple relation (4.7) is fulfilled or not. The assumption was that the probability that a decay is detected by one detector does not depend on whether the same decay is detected by the other detector or not.

The general idea of the β - γ -coincidence instrument is now (W. Bothe and H. J. v. Baeyer, 1935) that one detector should preferably be sensitive to the betas and one to the gammas. As we shall see in the description of the two detectors, this can only be achieved to some extent, but as it is our aim, we shall concentrate our interest on the independency of the emission of betas and gammas during the decay of Au^{198} .

The decay scheme of Au^{198} (fig. 5.1) includes three beta-branches, and it is seen that the highest-energy beta (from the transition directly to the ground state of Hg^{198}) is not followed by the emission of a gamma ray. If in the gamma detector we select the 411 keV gamma ray, we shall have a similar case with the part of the beta branch that goes through the 1087 keV level and from this directly to the ground state. Whether these two situations have any effect on the applicability of (4.7) depends on the dif-

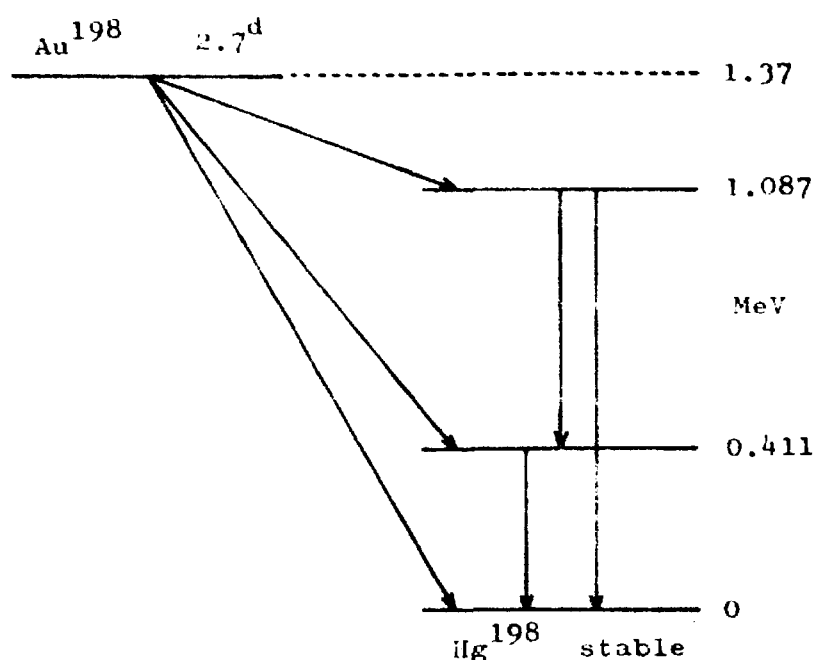


Fig. 5.1. Au^{198} decay scheme.

ferences in beta-detector effectiveness for the three beta-groups; with the beta detector in question these differences are small and go to zero when the thickness of the radioactive source becomes small.

In 99 per cent of the decays a beta group with maximum energy 959 keV is followed by a transition from the 411 keV level to the ground state in Hg^{198} . The direction of the gamma rays emitted in this case is correlated to that of the beta particles, the angular term depending on the energy of the beta particle (R. M. Steffen, 1960). Figure 5.2 shows this dependence, and it is possible to ensure by the design of the beta detector that it is isotropically sensitive to the higher-energy part of the beta group. We are therefore able to exclude any effect of the directional correlation.

The most serious complication in the decay of Au^{198} is the internal conversion of the 411 keV gammas. Nearly 3 per cent of the transition from the 411 keV level takes place through an emission of a conversion electron, and the beta detector is more sensitive to these electrons than to the betas if it is not already 100 per cent effective with respect to the latter.

With the above remarks in mind we must try to make the beta detector isotropically sensitive with as high an efficiency as possible. As

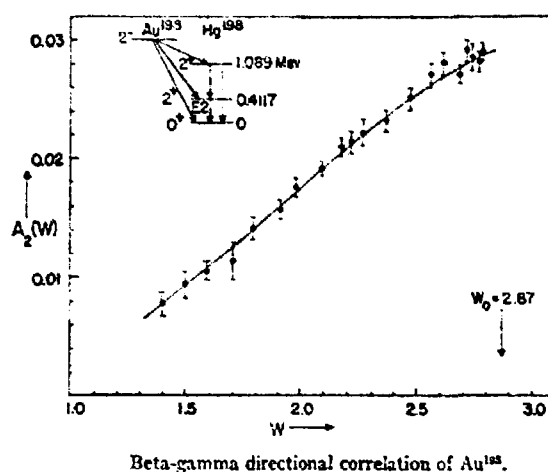


Fig. 5.2 (Steffen, 1960)

pointed out by O. M. Kofoed-Hansen at the Montreux conference (NBS Handbook 86, 1963), all terms correcting for branching contain the factor $(1 - \epsilon_\beta)/\epsilon_\beta$, which reduces the correction to a negligible fraction as the beta efficiency ϵ_β approaches unity.

A detailed calculation of these corrections in the case of Au^{198} has been reported by Wolfgang Pönitz (1963), but since there are small differences in the designs of the detectors, we arrange a series of measurements that enable us to fit Pönitz's result to the actual experiment. From our measurements, which will be described at the end of this chapter, we can conclude that we can use the correction calculated by Pönitz also in our experiment.

The Beta Detector

The beta detector consists of two flow proportional counters, one on each side of the radioactive sources. Figure 5.3 is a photograph of the detector when open. Some special details are:

- wire: stainless steel, diameter 0.1 mm;
- distance from wire to radioactive source: 3 mm;
- insulators: teflon;
- housing: gold-plated brass, right-angle box;
- source backing: $30 \mu\text{g}/\text{cm}^2$ VYNS film coated with $30 \mu\text{g}/\text{cm}^2$ gold;
- counting gas: flow of argon + 2 per cent methane;
- working voltage: 2200 V;
- multiplication factor: $10^4 - 10^5$.



Fig. 5.3. The beta detector. (Photo: Preben Nielsen)

The behaviour of the beta detector and the energy dissipation of the beta particles in the gas have recently been discussed by Leif Løvborg (1966); only some of his results will be mentioned here.

On account of the small size of the detector, the passage of a 1000 keV beta particle produces on an average approximately 100 primary ion pairs, corresponding to a mean energy loss of 2.6 keV. The mean energy loss of beta particles with lower primary energy will be greater, and in this way the primary beta spectrum is "inverted" into the electronic pulse-height spectrum. The low-energy beta particles are to be found in the group of large pulse heights and vice versa; furthermore the whole spectrum is smeared out because of scattering in the number of primary ion pairs.

The pulse-height spectrum from the beta detector is shown in figure 5.4. The most probable energy loss (the maximum point of the curve) is 4.4 keV in the case of Au^{198} , and for small pulse heights the spectrum curve climbs again owing to noise in the pre-amplifier and to multiple pulses arising from the impact of positive ions on the counter walls. These positive ions produce secondary electrons, which are delayed in time with respect to the passage of the beta particle; on gating of the multichannel analyser with signals from the gamma detector, these secondary impulses disappear. The discriminator in the beta channel is placed just at the minimum point of the ungated spectrum curve (fig. 5.4), and in the case of Au^{198} this discrimination level corresponds to a minimum energy loss of

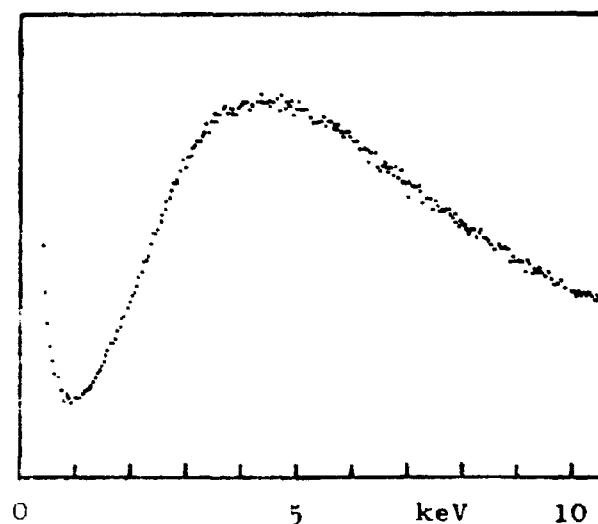


Fig. 5.4. The pulse-height spectrum from the beta detector.

0.7 keV or 30 ion pairs. The energy calibration was made by way of total absorption in the gas of characteristic X-rays from the decay of light elements.

The long tail of high pulses makes special demands on the electronic amplifiers, and we have used a double delay line amplifier with a conventional front-edge discriminator. A pre-amplifier close to the detector, giving an amplification of 5 times, will prevent serious noise take-up in the cable connecting the detector with the main amplifier.

We stated earlier by (4.18) that the efficiency of the beta detector, ϵ_{β} , was independent of r , θ and time (referring to figure 4.4). The time stability is achieved by routine checking of the amplifier gain, discriminator

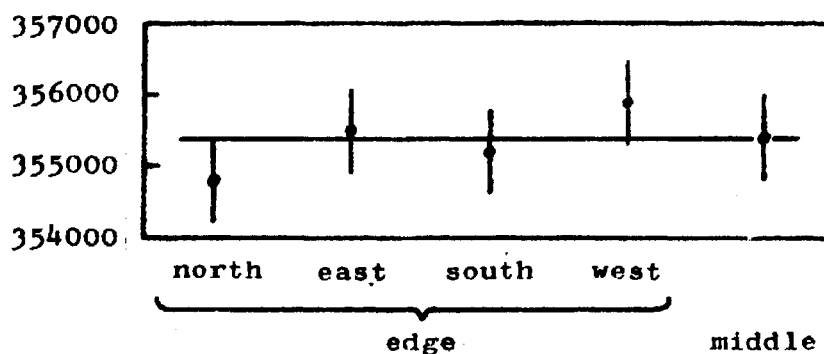


Fig. 5.5 Beta-detector sensitivity versus source position.

level and pulse-height distribution before and after each measurement. The independency of r and θ is demonstrated by a measurement with a gold foil (35 mg/cm^2 and 5.5 mm diameter), which was placed in five different positions, four at the edge of the counter (north, east, south, and west), and one in the middle of the counter. Figure 5.5 shows the results of these five measurements corrected for decay of the Au^{198} during the measurements.

The statement (4.18) is then seen to be amply fulfilled.

The Gamma Detector

The gamma detector consists of two thallium-activated sodium-iodine scintillation crystals, each 75 mm long by 75 mm diameter. They are placed coaxially with a distance of 44 mm between the outer cannings, from which it is approximately 8 mm to the active crystals. The mechanical design of the complete instrument is shown in the photograph figure 5.6 and in the drawing figure 5.7.

We claimed by (4.17) that the gamma-detection probability ϵ_γ should be independent of the z -direction (fig. 4.4), i. e. of displacements along the crystal axis. For each crystal we may assume a variation in detection probability inversely proportional to the square of the distance between the source and the detector surface, which is more unfavourable to our wishes than the actual distance law in the case of so big a crystal. Figure 5.8 shows the

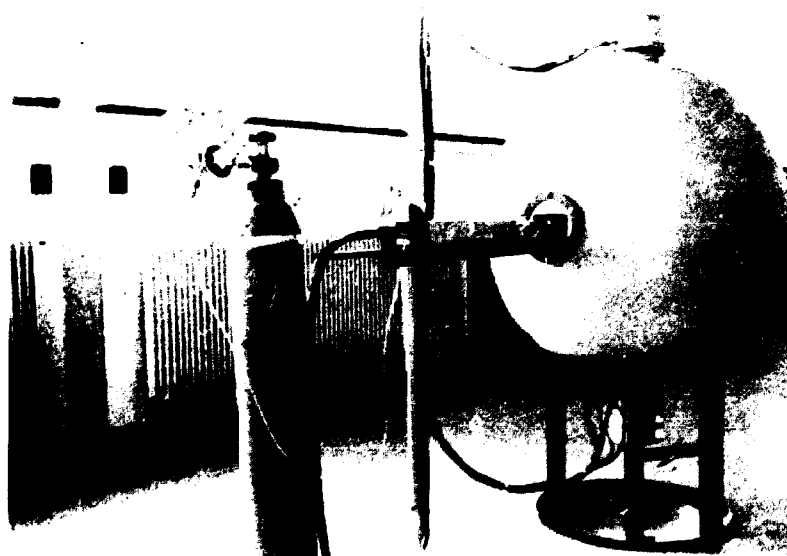


Fig. 5.6 Arrangement of detectors and shielding

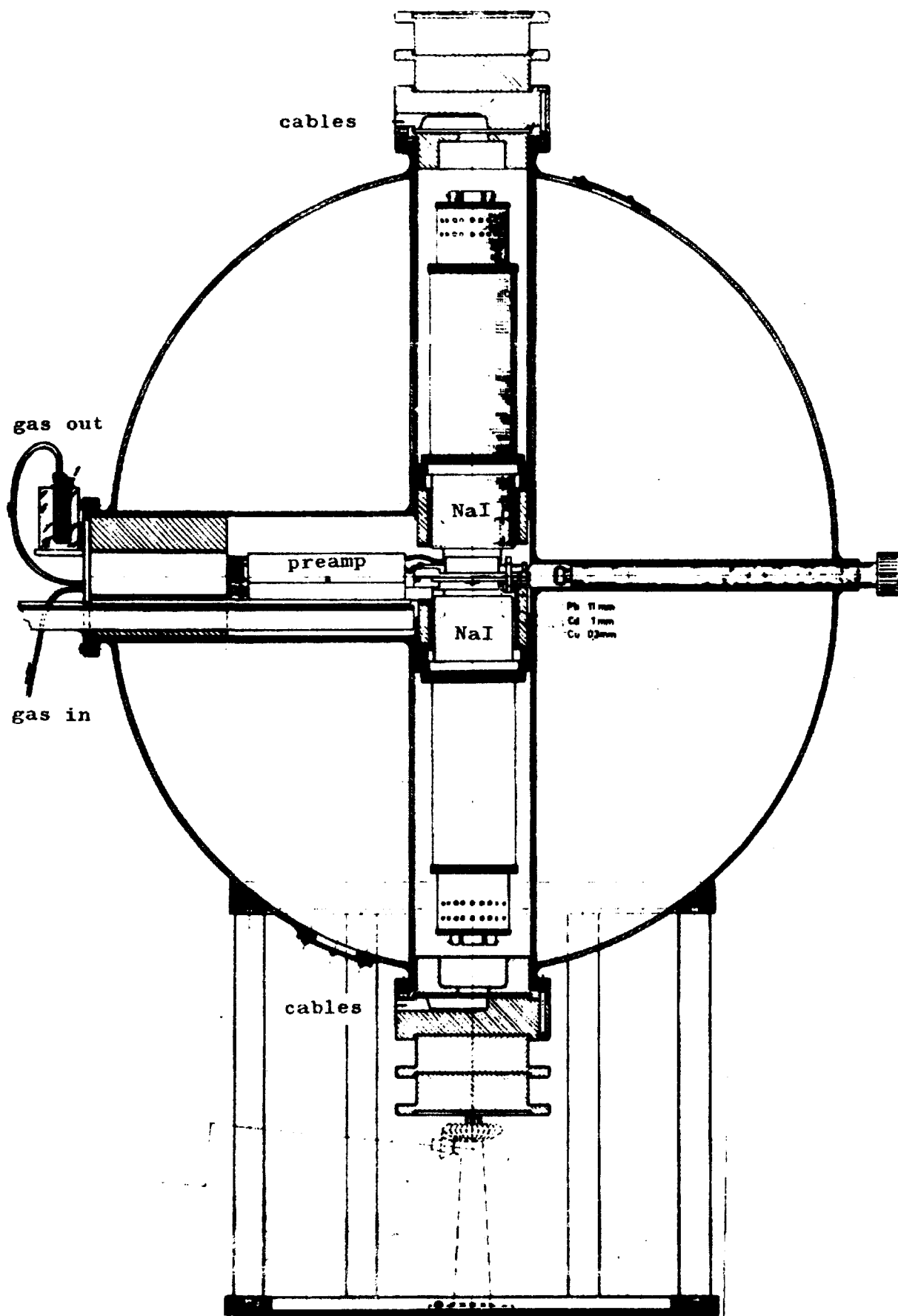


Fig. 5.7. Arrangement of detectors and shielding.

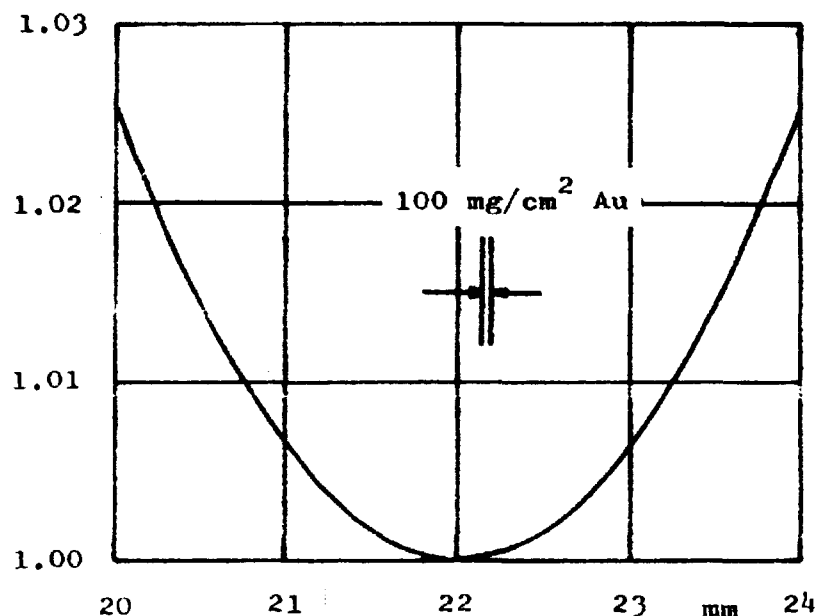


Fig. 5.8. Gamma detection probability versus z .

variation in detection probability when calculated on this basis. In the interval of 1 mm on both sides of the symmetry plane the biggest variation over an Au foil of thickness 75 mg/cm^2 will be 0.05 per cent, and since the foils are placed in this interval with an accuracy of less than 0.5 mm, the independency of the z -co-ordinate is proved.

The variation due to self-absorption in the foil is also negligible because of the symmetry in the design of the gamma detector, since the total detection probability for a gamma ray emitted at the height z above the mid-plane of a gold foil of thickness d contains the factor

$$e^{-\mu_0(\frac{d}{2} + z)} + e^{-\mu_0(\frac{d}{2} - z)} = 2 e^{-\mu_0 \frac{d}{2}} \cosh \mu_0 z, \quad (5.1)$$

where μ_0 is the total attenuation coefficient. Now z at its maximum is 30 mg/cm^2 , and μ_0 is for gold and for a gamma energy of 411 keV approximately $0.2 \text{ cm}^2/\text{g}$, so we have

$$\mu_0 \cdot z_{\max} = 0.2 \times 0.03 = 0.006. \quad (5.2)$$

Thus the total detection probability for radioactivity at the surface of the

foils is a factor

$$\cosh 0.006 = 1.00002 \quad (5.3)$$

greater than the corresponding figure for the mid-plane of the foils.

Each of the two scintillation detectors composing the gamma detector was equipped with an emitter follower, and the signals from these were added in a balancing circuit and passed to a double delay line amplifier and further to a single-channel analyser of the zero cross-over type. Before each measurement the position of the 411 keV line was found on the multi-channel analyser, and by means of an electronic pulse generator with high linearity the single-channel analyser was adjusted so that the relative positions of the line and the upper and lower discriminator level were kept constant.

The Electronics and the Coincidence Unit

A block diagram of the electronics is shown in figure 5.9. A more detailed diagram is not given since, even with fully transistorized equipment, it will be old at the date of printing and obsolete a few years later. Since the first report on an electronic coincidence unit (W. Bothe, 1929), coincidence units and other types of electronics have at any time been made of the best available components. Thus the only special feature is the de-

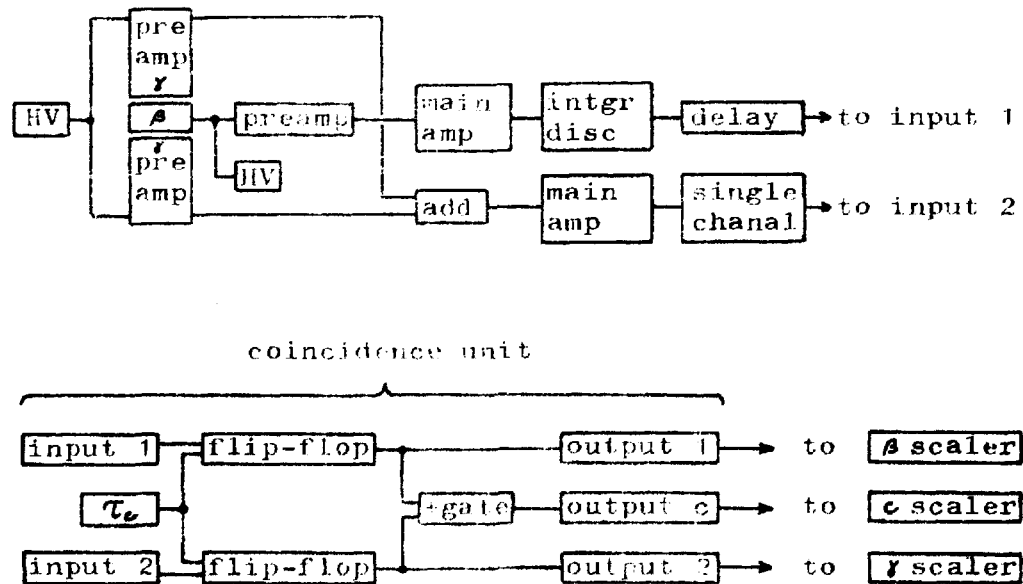


Fig. 5.9. Electronics.

sign of the coincidence unit, which should fulfil the requirements mentioned at the beginning of chapter 4.

In the design shown in fig. 5.9, simultaneous registrations in the beta and gamma scalers must necessarily cause a registration in the coincidence scaler too when the count rate is low. But if one of the detector impulses is delayed with respect to the other, both originating from the same decay, the coincidence registration may be lost if the resolving time is too short. A constant delay exists in our equipment since the discriminator in the beta branch is of the front-edge type and that in the gamma branch of the zero cross-over type. It is therefore advantageous to compensate for this by using a delay line in the beta branch so that the mean delay can be adjusted to zero. But time fluctuations in the beta detector can still give a time difference, and figure 5.10 shows the coincidence count-rates for different resolving times (1 and 2 μ s). It is seen that the mean delay corresponds to 35 scale divisions, which is approximately 0.4 μ s. In all subsequent measurements the delay was put at 35 scale divisions, and in

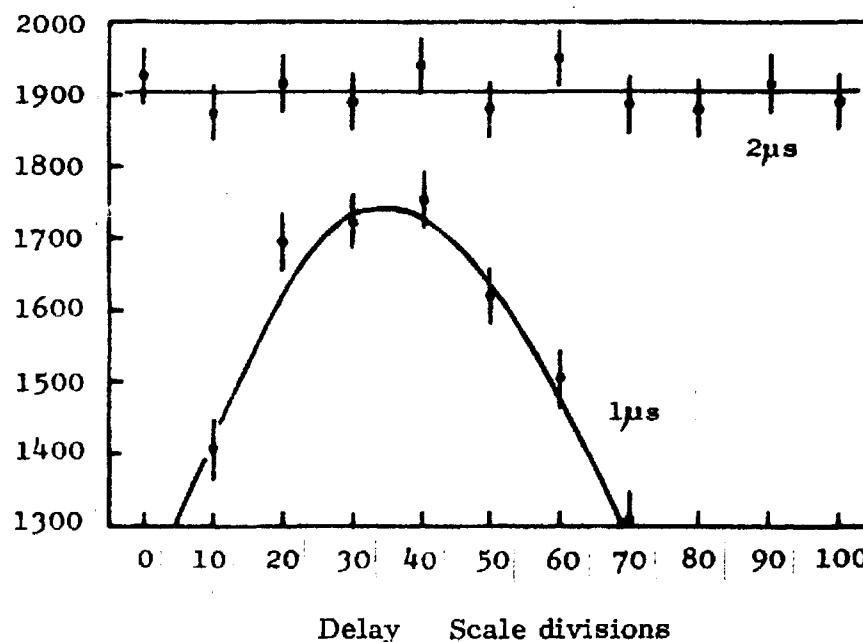


Fig. 5.10.

figure 5.11 the coincidence count-rate as a function of resolving time shows that if a resolving time of 3 μ s is adopted, the coincidence unit is 100 per cent effective.

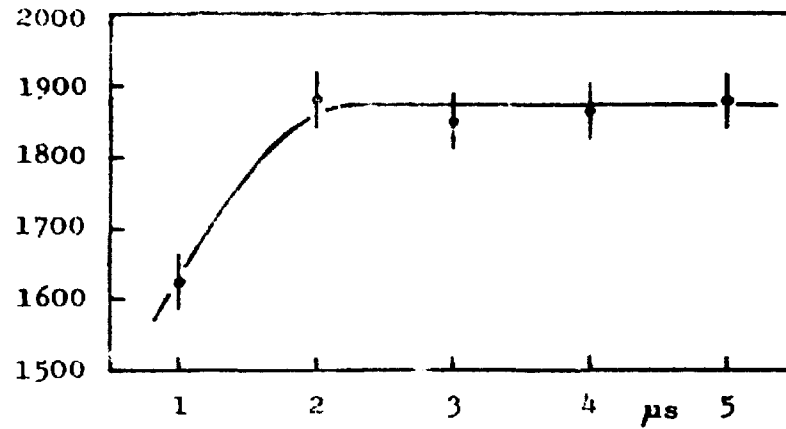


Fig. 5.11.

Corrections for Finite Foil Thickness

The above description of the details of the decay of Au^{198} and the instrument shows that all possible complications but one have been solved in the limit of low count-rates. The one which remains in this limit came mainly from the internal conversion of the 411 keV gamma ray. It was mentioned that even this effect vanishes in the limit of zero foil thickness, but when we use thicker foils, we must apply a correction factor.

Wolfgang Pönitz (1963) has reported calculations of this correction factor for an instrument resembling ours very much, but in order to fit these calculations to our instrument a series of measurements were made.

As an illustrative example, three foils (Nos. 1, 2 and 3), each 20 mg/cm^2 , were stacked and placed in front of a He^3 -counter in a neutron beam, where the upper end of the thermal and all the epithermal neutrons were filtered away by a 30 cm bismuth filter (J. Als-Nielsen, A. Bahnsen and W.K. Brown, 1966). After the irradiation the foils were counted both as single foils and sandwiched to simulate foils with thicknesses of 40 and 60 mg/cm^2 . After correcting for background (formula 6.20), count-rate effects (formulae 6.34 and 6.36) and decay, using the half-life $T_{1/2} = 2.7$ days, we found the count-rates of the first three foils to be

$N(1)$	$= 1082 \pm 1.7$	counts per second	
$N(2)$	$= 1070 \pm 1.7$	-	-
$N(3)$	$= 1046 \pm 1.7$	-	-
$N(1+2)$	$= 2184 \pm 3.2$	-	-
$N(2+3)$	$= 2144 \pm 3.6$	-	-
$N(1+3)$	$= 2154 \pm 2.7$	-	-
$N(1+2+3)$	$= 3281 \pm 6.7$	-	-

(5.4)

Through the correction factor $f(d)$ the absolute disintegration rate $N_0(1)$ is related to $N(1)$ by

$$N_0(1) = N(1) \cdot f(d) , \quad (5.5)$$

where d is the thickness of foil no. 1 (in this case 20 mg/cm^2).

The sum of the absolute disintegration rates, which must of course always be the same, can now be written as

$$\begin{aligned} N_0(1) + N_0(2) + N_0(3) &= \{ N(1) + N(2) + N(3) \} f(d) \\ &= \frac{1}{2} \{ N(1+2) + N(2+3) + N(1+3) \} f(2d) \\ &= N(1+2+3) f(3d) . \end{aligned} \quad (5.6)$$

The total correction factors $f'(d)$ as given by Pönitz (1963) are

$$\begin{aligned} f'(20 \text{ mg/cm}^2) &= 1 - 0.0148 = 1 - a_1 \\ f'(40 \text{ mg/cm}^2) &= 1 - 0.0264 = 1 - a_2 \\ f'(60 \text{ mg/cm}^2) &= 1 - 0.0349 = 1 - a_3 , \end{aligned} \quad (5.7)$$

but in order to cope with the said small differences in the experimental set-up we introduce a correction factor a , and so we write the corrected correction factor $f(d)$ as

$$f(d) = 1 - a \cdot a \quad (5.8)$$

and use our measurements to determine a . We thus have

$$N_1(1 - a a_1) = N_2(1 - a a_2) = N_3(1 - a a_3) , \quad (5.9)$$

where

$$\begin{aligned} N_1 &= N(1) + N(2) + N(3) &&= 3198 \pm 3.0 \\ N_2 &= \frac{1}{2} \{ N(1+2) + N(2+3) + N(1+3) \} &&= 3241 \pm 2.7 \\ N_3 &= N(1 + 2 + 3) &&= 3281 \pm 6.7 . \end{aligned} \quad (5.10)$$

Now (5.9) includes three possible equations with one unknown, and we can

regard (5.9) as the intersection of three lines of the form

$$y = N(1 - \alpha a) . \quad (5.11)$$

Figure 5.12 shows the three lines (5.9). Of course they do not intersect in the same point since N_1 , N_2 and N_3 are measured quantities with standard deviations σ_1 , σ_2 and σ_3 respectively.

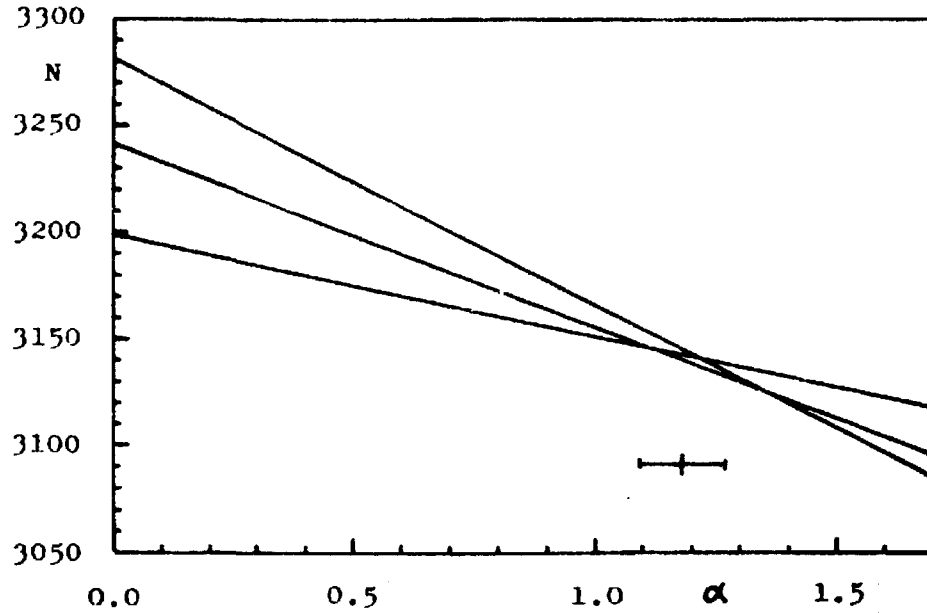


Fig. 5.12

Each line in figure 5.12 represents a probability distribution. The probability that line no. 1 "goes through" a square $d\alpha dy$ around the point (α, y) is given by

$$\frac{1}{\sigma_1 \sqrt{2\pi}} e^{-\frac{(y-N_1(1-\alpha a_1))^2}{2\sigma_1^2}} d\alpha dy , \quad (5.12)$$

and the probability, S , that all three lines cross that square is thus the product

$$S = \frac{1}{\sigma_1 \sigma_2 \sigma_3} \left(\frac{1}{\sqrt{2\pi}} \right)^3 e^{-\frac{1}{2} \left\{ \frac{(y-N_1(1-\alpha a_1))^2}{\sigma_1^2} + \frac{(y-N_2(1-\alpha a_2))^2}{\sigma_2^2} + \frac{(y-N_3(1-\alpha a_3))^2}{\sigma_3^2} \right\}}$$

$$S = c_1 e^{-\frac{1}{2} f(a, y)} da dy \quad (5.13)$$

Since we are only interested in a , we find the marginal distribution of a by integrating over the whole range of y , and we therefore make the rearrangement

$$\begin{aligned} f(a, y) &= \left\{ \frac{1}{\sigma_1^2} + \frac{1}{\sigma_2^2} + \frac{1}{\sigma_3^2} \right\} y^2 - 2 \left\{ \frac{N_1(1-aa_1)}{\sigma_1^2} + \frac{N_2(1-aa_2)}{\sigma_2^2} + \frac{N_3(1-aa_3)}{\sigma_3^2} \right\} y \\ &\quad + \frac{N_1^2(1-aa_1)^2}{\sigma_1^2} + \frac{N_2^2(1-aa_2)^2}{\sigma_2^2} + \frac{N_3^2(1-aa_3)^2}{\sigma_3^2} \\ &= \left\{ \frac{1}{\sigma_1^2} + \frac{1}{\sigma_2^2} + \frac{1}{\sigma_3^2} \right\} (y-y_0)^2 + \left\{ \frac{N_1^2(1-aa_1)^2}{\sigma_1^2} + \frac{N_2^2(1-aa_2)^2}{\sigma_2^2} + \frac{N_3^2(1-aa_3)^2}{\sigma_3^2} \right\} \\ &\quad - \frac{\left\{ \frac{N_1(1-aa_1)}{\sigma_1^2} + \frac{N_2(1-aa_2)}{\sigma_2^2} + \frac{N_3(1-aa_3)}{\sigma_3^2} \right\}^2}{\frac{1}{\sigma_1^2} + \frac{1}{\sigma_2^2} + \frac{1}{\sigma_3^2}} \end{aligned} \quad (5.14)$$

The probability distribution S is thus seen to be a Gaussian distribution in y for every fixed a , and therefore the integration over y from $-\infty$ to $+\infty$ will give

$$\int_{y=-\infty}^{y=+\infty} S(a, y) da dy = c_2 e^{-\frac{1}{2} h(a)} da, \quad (5.15)$$

where c_2 is a constant and $h(a)$ is given by

$$h(a) = \frac{N_1^2(1-aa_1)^2}{\sigma_1^2} + \frac{N_2^2(1-aa_2)^2}{\sigma_2^2} + \frac{N_3^2(1-aa_3)^2}{\sigma_3^2} -$$

$$- \frac{\frac{N_1(1-aa_1)}{\sigma_1^2} + \frac{N_2(1-aa_2)}{\sigma_2^2} + \frac{N_3(1-aa_3)}{\sigma_3^2}}{\frac{1}{\sigma_1^2} + \frac{1}{\sigma_2^2} + \frac{1}{\sigma_3^2}} . \quad (5.16)$$

By some tedious rearrangements of the same kind as in the derivation of (5.14) we find

$$h(a) = \frac{(a-a_0)^2}{\sigma_0^2} + \text{a constant} , \quad (5.17)$$

where

$$\frac{1}{\sigma_0^2} = \frac{N_1^2 a_1^2}{\sigma_1^2} + \frac{N_2^2 a_2^2}{\sigma_2^2} + \frac{N_3^2 a_3^2}{\sigma_3^2} - \frac{\left(\frac{N_1 a_1}{\sigma_1^2} + \frac{N_2 a_2}{\sigma_2^2} + \frac{N_3 a_3}{\sigma_3^2} \right)^2}{\frac{1}{\sigma_1^2} + \frac{1}{\sigma_2^2} + \frac{1}{\sigma_3^2}} \quad (5.18)$$

and

$$\frac{a_0}{\sigma_0^2} = \frac{N_1^2 a_1}{\sigma_1^2} + \frac{N_2^2 a_2}{\sigma_2^2} + \frac{N_3^2 a_3}{\sigma_3^2} - \frac{\left(\frac{N_1 a_1}{\sigma_1^2} + \frac{N_2 a_2}{\sigma_2^2} + \frac{N_3 a_3}{\sigma_3^2} \right) \left(\frac{N_1}{\sigma_1^2} + \frac{N_2}{\sigma_2^2} + \frac{N_3}{\sigma_3^2} \right)}{\frac{1}{\sigma_1^2} + \frac{1}{\sigma_2^2} + \frac{1}{\sigma_3^2}} . \quad (5.19)$$

We thus see that the marginal distribution is a Gaussian distribution in a with the mean value a_0 and the standard deviation σ_0 .

Figure 5.13 shows the background in the beta and the gamma detector, and table 5.1 gives the results of the measurements on two sets of three foils (20 mg/cm^2). The drop in gamma background between the 10th and 11th February, 1966, is caused by a corresponding drop in gamma detector efficiency as seen from table 5.1. Table 5.2 contains the results of one set of measurements on two 20 mg/cm^2 foils and one set of measurements on two 35 mg/cm^2 foils. For 35 and 70 mg/cm^2 foils Pönitz gives the theoretical values

$$f'(35 \text{ mg/cm}^2) = 1 - 0.0241 \quad (5.20)$$

and $f'(70 \text{ mg/cm}^2) = 1 - 0.0383$.

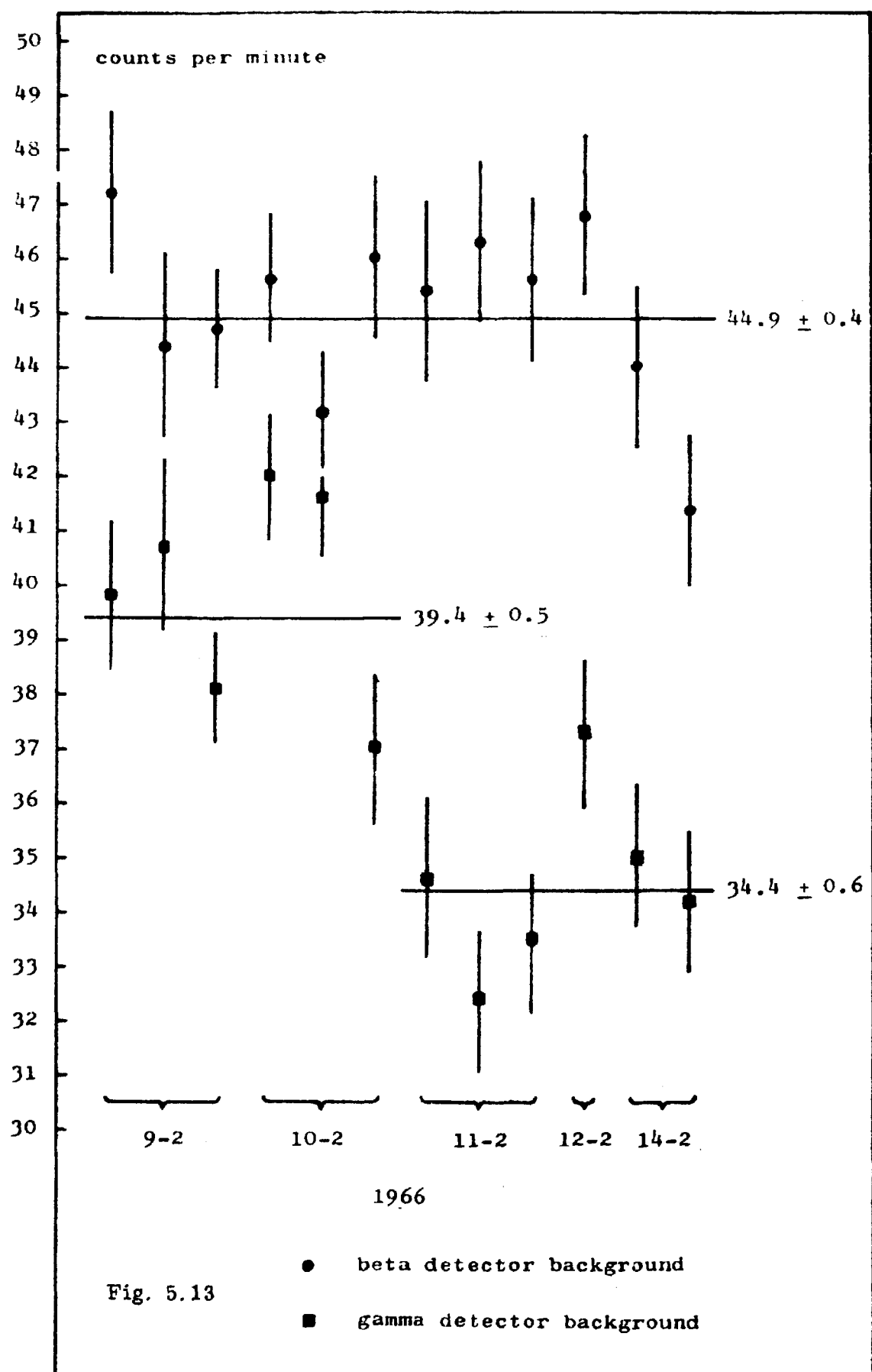


Fig. 5.13

Table 5.1

Foils i	Time 1966	E [β]	E [γ]	N	Count- rate correction	Decay correction	Final N(i)
(1)	9-2-14 ⁰⁰	0.738	0.115	1080.7 cps	+ 1.8 cps	1	1082 \pm 1.7 cps
(2)	9-2-15 ⁰⁰	0.735	0.115	1056.9 -	+ 1.7 -	1.0108	1070 \pm 1.7 -
(3)	9-2-16 ³⁶	0.738	0.116	1015.7 -	+ 1.6 -	1.0282	1046 \pm 1.7 -
(1+2)	10-2-9 ⁴⁵	0.600	0.115	1760.3 -	+ 7.2 -	1.2356	2184 \pm 3.2 -
(2+3)	10-2-10 ⁵¹	0.602	0.114	1708.9 -	+ 6.8 -	1.2498	2144 \pm 3.6 -
(1+3)	10-2-13 ²⁰	0.602	0.114	1668.6 -	+ 6.4 -	1.2858	2154 \pm 2.7 -
(1+2+3)	11-2-14 ⁵⁴	0.497	0.104	1933.7 -	+11.1 -	1.6872	3281 \pm 6.7 -
(4)	11-2-15 ⁵⁰	0.743	0.106	661.1 -	+ 0.7 -	1	662 \pm 1.4 -
(5)	11-2-16 ⁴⁵	0.743	0.106	634.1 -	+ 0.6 -	1.0098	642 \pm 1.4 -
(6)	12-2-10 ⁰²	0.741	0.105	532.0 -	+ 0.4 -	1.2150	647 \pm 1.4 -
(4+5)	14-2-15 ⁰⁶	0.606	0.106	615.1 -	+ 0.9 -	2.1432	1320 \pm 4 -
(5+6)	14-2-16 ⁰⁰	0.605	0.106	606.1 -	+ 0.9 -	2.1639	1313 \pm 4 -
(4+6)	14-2-16 ²⁸	0.602	0.106	612.6 -	+ 0.9 -	2.1747	1334 \pm 4 -
(4+5+6)	14-2-16 ⁵⁵	0.503	0.105	908.0 -	+ 2.4 -	2.1852	1989 \pm 5.7 -

Table 5.2

Foils i	Time 1966	$E[\epsilon_\beta]$	$E[\epsilon_\gamma]$	N	Count- rate correction	Decay correction	Final N(i)
(B1)	12-2-10 ⁵⁷	0.741	0.101	1095.4 cps	+ 1.8 cps	1	1097 \pm 2 cps
(B2)	12-2-12 ⁰⁷	0.741	0.101	1070.6 -	+ 1.8 -	1.0126	1086 \pm 2 -
(B1 + B2)	15-2-09 ⁴⁷	0.607	0.107	1028.0 -	+ 2.4 -	2.133	2198 \pm 2 -
1965							
(E1)	2-3-11 ²⁹	0.628	0.123	903 cps	+ 1.7 cps	1	905 \pm 2.4 cps
(E2)	2-3-12 ⁴⁰	0.631	0.124	948 -	+ 1.9 -	1.0127	962 \pm 2.2 -
(E1 + E2)	2-3-14 ²⁵	0.461	0.122	1821 -	+10.3 -	1.0319	1889 \pm 4.2 -

The calculations of the correction factor α gave

$$\begin{array}{ll}
 \text{from foils nos. 1, 2 and 3} & \alpha = 1.18 \pm 0.09 \\
 - \quad - \quad - \quad 4, 5 \text{ and } 6 & \alpha = 1.15 \pm 0.13 \\
 - \quad - \quad - \quad B1 \text{ and } B2 & \alpha = 0.58 \pm 0.16 \\
 - \quad - \quad - \quad E1 \text{ and } E2 & \alpha = 0.80 \pm 0.20 ,
 \end{array} \tag{5.21}$$

and the weighed mean (ref. III) of these four determinations is

$$\alpha = 1.04 \pm 0.06 . \tag{5.22}$$

We can thus conclude that we have confirmed the correction factor calculated by Pönitz (1963) to apply to our instrument within an accuracy given by the standard deviation of 6 per cent on the value of α . In the case of gold foils with a thickness of 20 mg/cm^2 the total correction factor

$$f(20 \text{ mg/cm}^2) = 0.9852 \tag{5.23}$$

is thus applicable with a standard deviation of

$$\sigma(f(20 \text{ mg/cm}^2)) = 0.0009 . \tag{5.24}$$

Comparison with the He^3 -Counter

As mentioned above, the three gold foils (e. g. nos. 1, 2 and 3) were irradiated stacked together in front of a He^3 -counter in order to standardize a neutron beam (J. Als-Nielsen, A. Bahnsen and W. K. Brown, 1966). The figure of interest is therefore the activity per mg/cm^2 extrapolated to the surface where the neutron beam leaves the gold foils.

Table 5.3 gives the results of the measurements on the two groups, each containing three foils, and the problem is now to find the probability distribution of the ordinate of a straight line fitting the three points at the abscissa t_x of the back surface. Let the line be

$$y = \alpha t + q . \tag{5.25}$$

We will then ask for the probability of

$$y_x = \alpha t_x + q \tag{5.26}$$

having a certain magnitude. The joint distribution $H(\alpha, q)$ of α and q is found in the next chapter to be

$$H(\alpha, q) = \frac{1}{\sigma_1 \cdots \sigma_n} \left(\frac{1}{\sqrt{2\pi}} \right)^n e^{-\frac{1}{2} f(\alpha, q)}, \quad (6.42)$$

where

$$f(\alpha, q) = A \alpha^2 + B q^2 + 2C \alpha q - 2D \alpha - 2E q + F, \quad (6.45)$$

and A, B, C, D, E , and F are given by (6.46), (6.47), (6.48), (6.49), (6.50), and (6.51) respectively.

We now eliminate α from (5.26) and (6.45) and find ($t_x \neq 0$)

$$\begin{aligned} f\left(\alpha = -\frac{q}{t_x} + \frac{y_x}{t_x}, q\right) &= A\left(-\frac{q}{t_x} + \frac{y_x}{t_x}\right)^2 + B q^2 + 2C\left(-\frac{q}{t_x} + \frac{y_x}{t_x}\right)q \\ &\quad - 2D\left(-\frac{q}{t_x} + \frac{y_x}{t_x}\right) - 2E q + F \\ &= \left(\frac{A}{t_x^2} + B - \frac{2C}{t_x}\right)q^2 - 2\left(\frac{A y_x}{t_x^2} - \frac{C y_x}{t_x} - \frac{D}{t_x} + E\right)q \\ &\quad + \frac{A}{t_x^2} y_x^2 - 2 \frac{D}{t_x} y_x + F \\ &= \left\{ \frac{A}{t_x^2} + B - \frac{2C}{t_x} \right\} \left\{ q - \frac{\frac{A}{t_x^2} t_x - \frac{C}{t_x} y_x - \frac{D}{t_x} + E}{\frac{A}{t_x^2} + B - \frac{2C}{t_x}} \right\}^2 \\ &\quad - \frac{\left(\frac{A}{t_x^2} y_x - \frac{C}{t_x} y_x - \frac{D}{t_x} + E \right)^2}{\frac{A}{t_x^2} + B - \frac{2C}{t_x}} + \frac{A}{t_x^2} y_x^2 - 2 \frac{D}{t_x} y_x + F. \end{aligned} \quad (5.27)$$

As we must take any possible values of q into account, we insert (5.27) in (6.42) and integrate over q from $-\infty$ to $+\infty$; we find

$$H(y_x) = \int_{-\infty}^{\infty} H\left(\alpha = -\frac{q}{t_x} + \frac{y_x}{t_x}, q\right) dq = (\text{a constant}) \cdot e^{-\frac{1}{2} f(y_x)}, \quad (5.28)$$

Table 5.3

Foil no.	Thick- ness mg/cm ²	Midpoint t mg/cm ²	Activity per thickness + σ counts per second per mg/cm ²
(1)	20.065	-20.116	53.95 \pm 0.09
(2)	20.168	0	53.05 \pm 0.09
(3)	20.018	+20.093	52.25 \pm 0.09
Back surface		30.102	
(4)	20.088	-19.876	32.94 \pm 0.07
(5)	19.665	0	32.64 \pm 0.07
(6)	20.080	+19.872	32.21 \pm 0.07
Back surface		+29.912	

where

$$\begin{aligned}
 f(y_x) &= - \frac{\left(\frac{A}{t_x^2} - \frac{C}{t_x}\right)^2}{\frac{A}{t_x^2} + B - \frac{2C}{t_x}} y_x^2 + 2 \frac{\left(\frac{A}{t_x^2} - \frac{C}{t_x}\right)\left(\frac{D}{t_x} - E\right)}{\frac{A}{t_x^2} + B - \frac{2C}{t_x}} y_x \\
 &\quad + \frac{A}{t_x^2} y_x^2 - 2 \frac{D}{t_x} y_x \\
 &= \left\{ \frac{A}{t_x^2} - \frac{\left(\frac{A}{t_x^2} - \frac{C}{t_x}\right)^2}{\frac{A}{t_x^2} + B - \frac{2C}{t_x}} \right\} y_x^2 - 2 \left\{ \frac{D}{t_x} + \frac{\left(\frac{A}{t_x^2} - \frac{C}{t_x}\right)\left(E - \frac{D}{t_x}\right)}{\frac{A}{t_x^2} + B - \frac{2C}{t_x}} \right\} y_x \\
 &= \frac{(y_x - y_0)^2}{\sigma^2(y_0)} + \text{a constant.} \tag{5.29}
 \end{aligned}$$

The expression (5.29) shows that the probability distribution of y_x is a

Gaussian distribution with the mean value y_0 and the standard deviation $\sigma(y_0)$ given by

$$\frac{y_0}{\sigma^2(y_0)} = \frac{D}{t_x} + \frac{\left(\frac{A}{t_x^2} - \frac{C}{t_x}\right)\left(E - \frac{D}{t_x}\right)}{\frac{A}{t_x^2} + B - \frac{2C}{t_x}} \quad (5.30)$$

and

$$\frac{1}{\sigma^2(y_0)} = \frac{A}{t_x^2} - \frac{\left(\frac{A}{t_x^2} - \frac{C}{t_x}\right)}{\frac{A}{t_x^2} + B - \frac{2C}{t_x}} = \frac{\frac{A}{t_x^2} \cdot B - \left(\frac{C}{t_x}\right)^2}{\frac{A}{t_x^2} + B - \frac{2C}{t_x}} \quad (5.31)$$

Figure 5.14 shows the numerical results based on the values in table 5.3. The values for the slope were found to be

$$a_{1,2,3} = (80 \pm 6) \cdot 10^{-5} \text{ cm}^2/\text{mg} \quad (5.32)$$

$$a_{4,5,6} = (57 \pm 3) \cdot 10^{-5} \text{ cm}^2/\text{mg} \quad (5.33)$$

with the mean value

$$a = (72 \pm 5) \cdot 10^{-5} \text{ cm}^2/\text{mg} \quad (5.34)$$

Since 1 mg gold contains

$$\frac{10^{-3}}{197.2} \cdot 6.022 \cdot 10^{23} = 3.054 \cdot 10^{18} \text{ atoms}, \quad (5.35)$$

the attenuation per atom is

$$a = \frac{(72 \pm 5) \cdot 10^{-5}}{3.054 \cdot 10^{18}} \text{ cm}^2 = (236 \pm 17) \cdot 10^{-24} \text{ cm}^2 \quad (5.36)$$

Comparing this value with the thermal absorption cross section for gold of

$$\sigma_{\text{abs}}(2200 \text{ m/s}) \sim 98 \cdot 10^{24} \text{ cm}^2/\text{atom}, \quad (5.37)$$

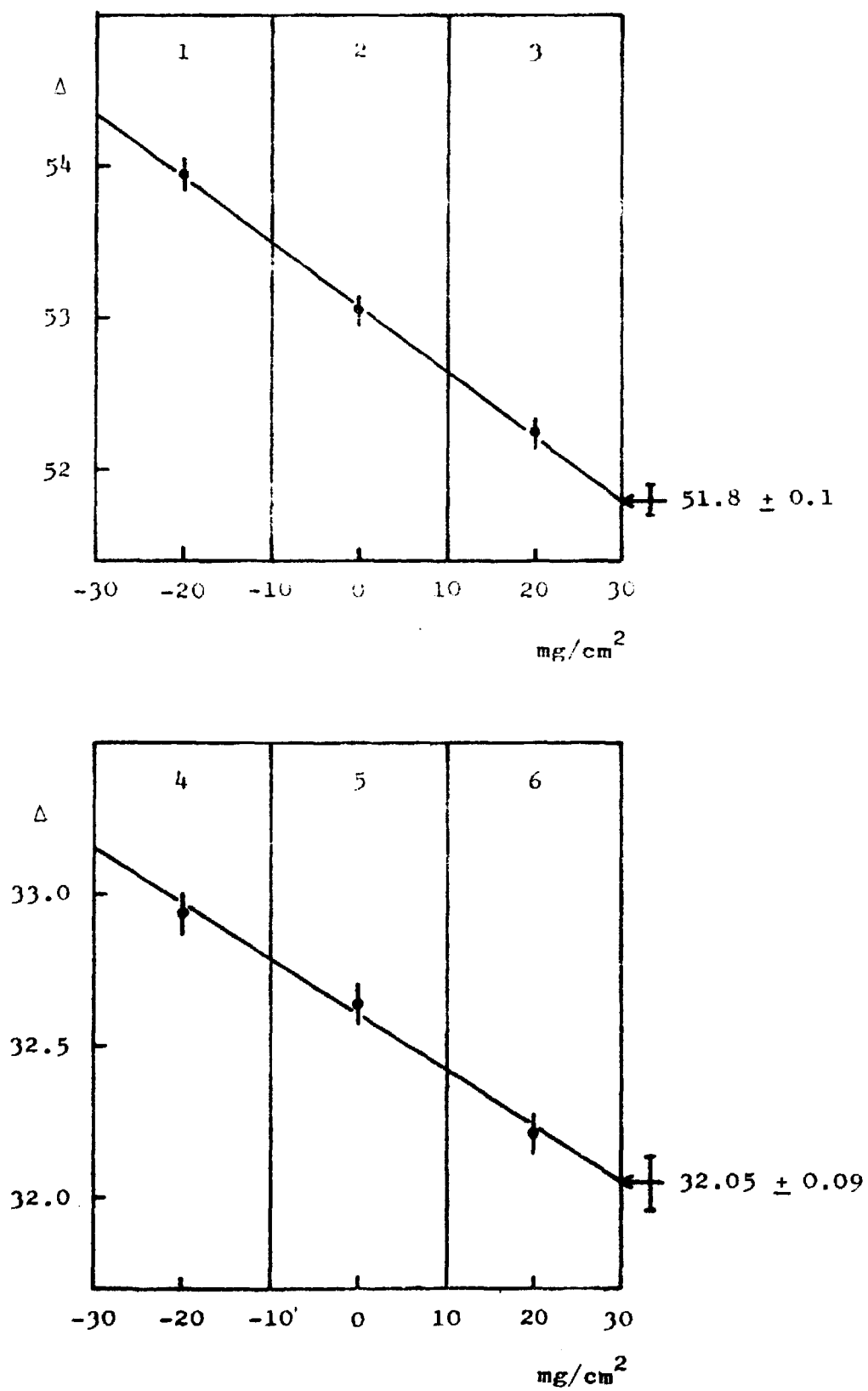


Fig. 5.14. Extrapolation to the back of the foils.

we can conclude that, after filtration with the above-mentioned Bi-filter, the neutron beam consists of rather "cold" neutrons. However, since the neutron spectrum is far from a Maxwellian spectrum, it is not possible to define a temperature that could be assigned to the neutrons.

For the standardization of the neutron beam with respect to the neutron density, the attenuation cross section just found is not needed since it is only necessary to introduce a small correction for gold for not having a perfect $1/v$ cross section. With these corrections the determination of the neutron density in the beam based on the measurements of gold foils agrees with determinations based on measurements with a H^3 proportional counter within the precision of both instruments (J. Als-Nielsen, A. Bahnsen and W. K. Brown, 1966).

CHAPTER 6:

COUNT-RATE-DEPENDENT CORRECTIONS

One of our fundamental hypotheses on which the evaluations in the foregoing chapters are based was that the probability that a decay will occur and be detected in a time interval dt is proportional to dt and independent of any other decays or registrations.

We thus found (ref. II) that the probability that the first decay which occurs and is registered in the time interval from t to $t+dt$ after the time $t=0$ is given by

$$G(0,1; t)dt = a \cdot e^{-at} dt, \quad (6.1)$$

since

$$e^{-at} = 1 - (1 - e^{-at}) \quad (6.2)$$

is the probability of getting no counts in the interval from 0 to t , and

$$adt \quad (6.3)$$

is the probability of getting a registration in the interval dt .

In normal electronic equipment, however, the registration of a decay will make any further registrations impossible for a certain period of time. For the sake of argument let us assume that the period in which the equipment is unable to make such further registrations is a constant τ . This means that a registration at the time t_0 has made the equipment insensitive

until the time $t_0 + \tau$. The probability that registration number two will occur between the times $t \geq t_0 + \tau$ and $t + dt$ is again given by

$$G(1, 2; t)dt = a e^{-a(t-(t_0 + \tau))} dt \quad (6.4)$$

since the equipment was assumed to be fully sensitive after the time $t_0 + \tau$.

Thus the probability that the second registration will take place between $t \geq \tau$ and $t + dt$ independently of the time t_0 of the first registration, is given as

$$\begin{aligned} G(0, 2; t)dt &= \int_{t_0=0}^{t_0=t-\tau} G(0, 1; t_0)dt_0 G(1, 2; t)dt \\ &= \int_{t_0=0}^{t_0=t-\tau} a e^{-at_0} dt_0 a e^{-a(t-(t_0 + \tau))} dt \\ &= a^2 e^{-a(t-\tau)} dt \int_{t_0=0}^{t_0=t-\tau} dt_0 \\ &= a^2 e^{-a(t-\tau)} (t-\tau) dt. \end{aligned} \quad (6.5)$$

If the second registration comes at t_1 , the probability of the third coming between $t \geq t_1 + \tau$ and $t + dt$ will be

$$G(2, 3; t)dt = a e^{-a(t-(t_1 + \tau))} dt; \quad (6.6)$$

thus, by integrating over all possible values of t_1 , we find

$$\begin{aligned}
G(0, 3; t) dt &= \int_{t_1 = \tau}^{t_1 = t - \tau} G(0, 2; t_1) dt_1 G(2, 3; t) dt \\
&= \int_{t_1 = \tau}^{t_1 = t - \tau} a^2 e^{-a(t_1 - \tau)} (t_1 - \tau) dt_1 a e^{-a(t - (t_1 + \tau))} dt \\
&= a^3 e^{-a(t - 2\tau)} dt \int_{t_1 = \tau}^{t_1 = t - \tau} (t_1 - \tau) dt_1 \\
&= a^3 e^{-a(t - 2\tau)} \frac{1}{2} (t - 2\tau)^2 dt. \tag{6.7}
\end{aligned}$$

From (6.7) we guess that the probability of having the $n + 1^{\text{st}}$ registration between $t \geq n\tau$ and $t + dt$ is given by

$$G(0, n+1; t) dt = a^{n+1} e^{-a(t - n\tau)} \frac{1}{n!} (t - n\tau)^n dt. \tag{6.8}$$

If this is true, we find

$$\begin{aligned}
G(0, n+2; t) dt &= \int_{t_0 = n\tau}^{t_0 = t - \tau} a^{n+1} e^{-a(t_0 - n\tau)} \frac{1}{n!} (t_0 - n\tau)^n dt_0 a e^{-a(t - (t_0 + \tau))} dt \\
&= a^{n+2} e^{-a(t - (n+1)\tau)} \frac{dt}{n!} \int_{t_0 = n\tau}^{t_0 = t - \tau} (t_0 - n\tau)^n dt_0 \\
&= a^{n+2} e^{-a(t - (n+1)\tau)} \frac{1}{(n+1)!} (t - (n+1)\tau)^{n+1} dt \tag{6.9}
\end{aligned}$$

We are thus able to prove the expression (6.8) by induction.

From the interval distribution (6.8) the probability of obtaining just n registrations in the interval from 0 to T is found by stating that the n^{th} registration should take place before T and the $n + 1^{\text{st}}$ registration after T .

Now there exist only three possible, mutually excluding events:

- (1) the n^{th} registration takes place after T with the probability

$$\int_T^{\infty} G(0, n; t) dt ; \quad (6.10)$$

- (2) the n^{th} registration takes place before T and the $n + 1^{\text{st}}$ registration after T with the probability

$$G(0, T, a, \tau ; n) ; \quad (6.11)$$

- (3) the $n + 1^{\text{st}}$ registration takes place before T with the probability

$$1 - \int_T^{\infty} G(0, n+1 ; t) dt . \quad (6.12)$$

Since the summation of these three probabilities should give unity, we find

$$\begin{aligned} G(0, T, a, \tau ; n) &= \int_T^{\infty} G(0, n+1 ; t) dt - \int_T^{\infty} G(0, n; t) dt \\ &= \int_T^{\infty} a^{n+1} e^{-a(t-n\tau)} \frac{(t-n\tau)^n}{n!} dt \end{aligned} \quad (6.13)$$

$$- \int_T^{\infty} a^n e^{-a(t-(n-1)\tau)} \frac{(t-(n-1)\tau)^{n-1}}{(n-1)!} dt$$

Only in the limit $\tau \rightarrow 0$ is the probability of observing n registrations in the time interval from 0 to T given by the Poisson distribution, i. e.

$$G(0, T, a, \tau; n) \rightarrow \frac{(aT)^n}{n!} e^{-aT} \quad \text{as} \quad \tau \rightarrow 0, \quad (6.14)$$

which can be proved by using the formula

$$\int_0^\infty \frac{x^n}{n!} e^{-x} dx = \sum_{i=0}^n \frac{x^i}{i!} e^{-x}. \quad (6.15)$$

These results were for the simplest possible assumptions with a single detector; in the case of coincidence equipment very special designs must be made in order to give a rigorous description of the dead-time correction (Westcott, 1948; Westcott, Greenberg, Kirkaldy, 1953; Gandy, 1962).

The time interval until the first registration takes place is not affected by the time the equipment requires to reach the initial condition. If both channels are closed, whatever the type of registrations has been, then the total time the instrument has been open will be the correct counting time. The correction can be made automatically in the so-called life-time integrator (Gandy, 1963).

But there are other corrections to be made, and the question remains whether the proper action of the electronics depends on the count-rate. The problem can be solved experimentally since it is possible to find the total correction due to count-rate, of whatever composite nature it may be, by the proper use of the substitution method (ref. 1).

The Substitution Method in the Case of Au^{198}

In the case of Au^{198} , which has a half-life of 2.7 days, it is necessary to modify the procedure described for radionuclides with very long half-lives (ref. 1). It was shown that if the ratio between the activities of successive radioactive sources in a set was

$$1 : 1.618, \quad (6.16)$$

it should suffice to have two sources in the detector at a time, and with a

radionuclide with a half-life of 2.7 days this ratio is achieved after a decay of 45 hours. So if we start with two sources with the activity ratio (6.16) and make observations every second day, these two sources will simulate a set with a large number of sources. If we want to cover an activity scale of 2 decades, we must have a set with nine sources, and this is simulated by measuring the same two gold foils over a period of sixteen days.

It is therefore necessary to have a procedure as described in chapter 5, which can control the stability of the instrument, but if we are to believe the applicability of corrections, we should demand this stability anyhow.

In order to facilitate the calculations, a standardized procedure was adopted. After controlling the whole experimental set-up in the manner described in chapter 5, the following observations were made with the counting time fixed at 100 sec and the repetition time fixed at 120 sec:

$$\begin{aligned}
 10^h \ 0^m: & \quad 5 \text{ times background } b_{\beta I}, b_{\gamma I}, b_{cI} \\
 10^h \ 15^m: & \quad 5 \text{ times foil no. 1 } n_{\beta I}, n_{\gamma I}, n_{cI} \\
 10^h \ 30^m: & \quad 5 \text{ times foil no. 1 + foil no. 2 } n_{\beta I+2}, n_{\gamma I+2}, n_{cI+2} \\
 10^h \ 45^m: & \quad 5 \text{ times foil no. 2 } n_{\beta 2}, n_{\gamma 2}, n_{c2} \\
 11^h \ 0^m: & \quad 5 \text{ times background } b_{\beta 2}, b_{\gamma 2}, b_{c2} .
 \end{aligned}$$

This schedule allows for exchange of the foils and flowing with the beta counter gas before each new measurement.

Each single number in the scheme above, e. g. $n_{\beta I}$, is thus the summation of the count numbers for five periods of 100 sec each, and hence stands for the total number of registrations for a period of 500 sec. The division of the observation into five periods gives a hint of whether the detectors are working properly (ref. II, p. 14).

Division of the count-numbers by the observation time, t , gives the corresponding count-rates, e. g.

$$N_{\beta I} = \frac{n_{\beta I}}{t} . \quad (6.17)$$

Subtraction of Background

Our description of the statistics of the experiment was based on the independent statistical variables n_1 , n_2 and n_3 connected with the count numbers n_{β} , n_{γ} and n_c by (4.4). In the same way we can define the statistically independent background count numbers

$$\begin{aligned}
b_1 &= b_c \\
b_2 &= b_\beta - b_c \\
b_3 &= b_\gamma - b_c ,
\end{aligned} \tag{6.18}$$

and if the efficiency of the beta detector approaches unity, the number n_3 of gammas which are not coincident with the betas will vanish, i. e. n_3 will be small or comparable with b_3 . It will then be necessary to estimate the true number of non-coincident gammas n_3 according to reference II, and doing so, we shall never have a beta-detector effectiveness

$$E[\epsilon_\beta] = \frac{n_1}{n_1 + n_3} \tag{6.19}$$

greater than unity.

It is desirable to keep b_3 as low as possible, and this is here done, as shown in figure 5.7, by surrounding the detectors with a sea-mine shell filled with small steel pellets, giving a shell of approximately 30 cm steel.

In our case, where the beta-detector effectiveness is far from unity, n_3 is much greater than b_3 ; therefore we calculate the number

$$\begin{aligned}
N_i &= \frac{(N_{i\beta} - B_\beta)(N_{i\gamma} - B_\gamma)}{N_{ic} - B_c} \\
&= \frac{1}{t} \cdot \frac{(n_{i\beta} - b_\beta)(n_{i\gamma} - b_\gamma)}{(n_{ic} - b_c)} .
\end{aligned} \tag{6.20}$$

and, except for the correction due to the decay scheme (chapter 5) and a count-rate-dependent correction, N_i is the absolute decay rate.

When the count-rates become higher, it is in fact not correct to subtract the background in this manner since the background is measured at a low and the activity plus the background at a high count-rate (Campion, 1959). The error introduced in this way is thus dependent on the count-rate, and here we will just pool it with all the other count-rate-dependent errors.

We can now proceed with a formalistic definition of the count-rate-dependent correction by writing

$$A_i = N_i f(N_i) , \quad (6.21)$$

where N_i is the "observed" count-rate (ref. I) calculated by means of (6.20), and A is the corresponding "true" count-rate free from count-rate-dependent corrections. We further know that

$$f(N_i) \rightarrow 1 \quad \text{as} \quad N_i \rightarrow 0 \quad (6.22)$$

(ref. I).

If we do not have too high count-rates, we may use a linear approximation to $f(N_i)$ by writing

$$A_i = N_i(1 + \tau N_i) , \quad (6.23)$$

where τ is an empirical parameter that has the dimension of time and must be determined by an appropriate method (Thomas 1962). Here we will use the substitution method to determine $f(N)$, and except for the limit given by (6.22) it is not necessary to assume a linear correction of the form (6.23).

From each day's measurements we then calculate N_I , N_{I2} and N_2 , and by (6.21) we have

$$\begin{aligned} A_I &= N_I f(N_I) \\ A_{I2} &= N_{I2} f(N_{I2}) \\ A_2 &= N_2 f(N_2) . \end{aligned} \quad (6.24)$$

Since A_{I2} is the true count-rate when foils nos. 1 and 2 are placed alongside each other in the counter, and since we have demonstrated that the instrument was insensitive to r and θ (figures 4.4 and 5.5), the difference between A_{I2} and $A_I + A_2$ will be caused only by the decay in source strength during the measurements, and thus we have

$$A_{I2} = A_I \cdot e^{-\lambda t_0} + A_2 \cdot e^{+\lambda t_0} , \quad (6.25)$$

where λ is the decay constant of Au^{198} and $t_0 = 15$ min is the time between the measurements. Using the half-life $T_{1/2} = 2.7$ days, we find

$$e^{+\lambda t_0} = 1.0027 \quad (6.26)$$

$$\text{and } e^{-\lambda t_0} = 0.9973 . \quad (6.27)$$

Since these corrections are small, and $f(N)$ is a slowly varying function of N , we will just correct the observed numbers by

$$N'_1 = N_1 \cdot 0.9973 \quad (6.28)$$

$$\text{and } N'_2 = N_2 \cdot 1.0027 .$$

Using (6.24) and (6.25), we find

$$N_{12} f(N_{12}) = N'_1 f(N'_1) + N'_2 f(N'_2) \approx N'_1 f(N'_1) + N'_2 f(N'_2) , \quad (6.29)$$

which is just formula (9) in reference I. If at a later stage it should be clear that one or more of our approximations have been too rough, we must go back to this point, and thus equation (6.29) should be taken only as the first step (and perhaps the last too) of an iteration.

From (6.29) we find

$$f(N_{12}) = \frac{1}{N_{12}} \left\{ N'_1 f(N'_1) + N'_2 f(N'_2) \right\} , \quad (6.30)$$

which expresses the correction factor at the observed count-rate N_{12} by the correction factor at the two lower count-rates N'_1 and N'_2 .

The observations are given in table 6.1, where the directly observed count numbers have been corrected for background counts by (6.20) and for decay during the observation time by (6.28). The standard deviations have been calculated from (4.44). The reference time is the beginning of the counting period.

After the first three measurements (foils nos. I and II) it was clear that the neutron irradiation, which should have produced an activity ratio given by (6.16), had not been completely successful, and it was then decided to lower the activity of foil no. II by cutting away a piece of it. After this operation the name was changed from foil no. II to foil no. 2.

Table 6.1 also includes the ratios N_1'/N_{12} and N_2'/N_{12} and finally $f(N_{12})$ calculated from formula (6.30).

The calculations of $f(N_{12})$ start at the bottom of table 6.1 on the assumption

$$f(106) = f(164) = 1, \quad (6.31)$$

and the standard deviations assigned to the calculated $f(N)$ account only for the standard deviations of the count-rates N_1' , N_{12} and N_2' in formula (6.30) and not for those of $f(N_1')$ or $f(N_2')$. In this way we see the standard deviations of the calculated $f(N)$ relative to the vicinity. The total standard deviations, which are to some extent accumulated from all lower measurements (ref. 1), will be strongly correlated.

Figure 6.1 shows these calculated values of $f(N)$, and it is seen that a linear approximation is possible up to a count-rate of 10 000 counts per second. A straight line fitting the points does not go through the point $(N, f(N)) = (0, 1)$, but this is due to the large standard deviations for the lowest count-rates. By extrapolating the line to $N = 0$, and demanding (6.22) to be fulfilled, we find the correct $f(N)$. By the use of formulae (6.53) and (6.54), which will be developed below, we find the parameter τ in the linear expression (6.23) to be

$$\tau = 2.1 \pm 0.2 \mu s. \quad (6.32)$$

This value of τ is easily explained by the design of the instrument, all time constants (pulse widths, resolving times in scalers, etc.) being small (below 0.5 μs) except the coincidence resolving time τ_c , which was 3 μs with an accuracy of $\pm 10\%$. The first count-rate effect to be expected will therefore be random coincidences, and the ratio of the number of random to the number of true coincidences is

$$\frac{(N_c)_{\text{random}}}{(N_c)_{\text{true}}} = 2 \tau_c (1 - \epsilon_\beta)(1 - \epsilon_\gamma) N \quad (6.33)$$

(Evans, 1955). Accordingly the count-rate-dependent correction should be

Table 6.1

Ref. time 1965	N_I' cps	N_{II} cps	N_{II}' cps	N_I'/N_{II}	N_{II}'/N_{II}	$f(N_{II})$
24 Mar 11 ^h 15 ^m	6228 ± 8	1173 ±	11802 ± 10	5.31	10.06	- ± -
26 Mar 10 ^h 15 ^m	3794 ± 6	10869 ± 10	7195 ± 8	.3491	.6620	1.044 ± 0.0015
28 Mar 11 ^h 15 ^m	2253 ± 5	6507 ± 8	4278 ± 7	.3462	.6574	1.033 ± 0.002
	N_I' cps	N_{I2} cps	N_2' cps	N_I'/N_{I2}	N_2/N_{I2}	$f(N_{I2})$
28 Mar 12 ^h 45 ^m	2214 ± 5	5631 ± 8	3446 ± 6	.3932	.6120	1.033 ± 0.002
30 Mar 10 ^h 15 ^h	1359 ± 4	3468 ± 6	2127 ± 5	.3919	.6133	1.030 ± 0.0025
2 Apr 10 ^h 15 ^m	816 ± 3	2086 ± 5	1273 ± 4	.3912	.6102	1.025 ± 0.003
4 Apr 10 ^h 15 ^m	489 ± 2	1247 ± 4	764 ± 3	.3921	.6127	1.025 ± 0.004
6 Apr 10 ^h 15 ^m	293 ± 1.6	744 ± 3	454 ± 2	.3938	.6102	1.021 ± 0.005
8 Apr 10 ^h 15 ^m	175 ± 1.3	444 ± 2	273 ± 1.6	.3941	.6149	1.018 ± 0.007
10 Apr 10 ^h 15 ^m	106 ± 1	266 ± 1.6	164 ± 1.3	.3925	.6165	1.015 ± 0.009

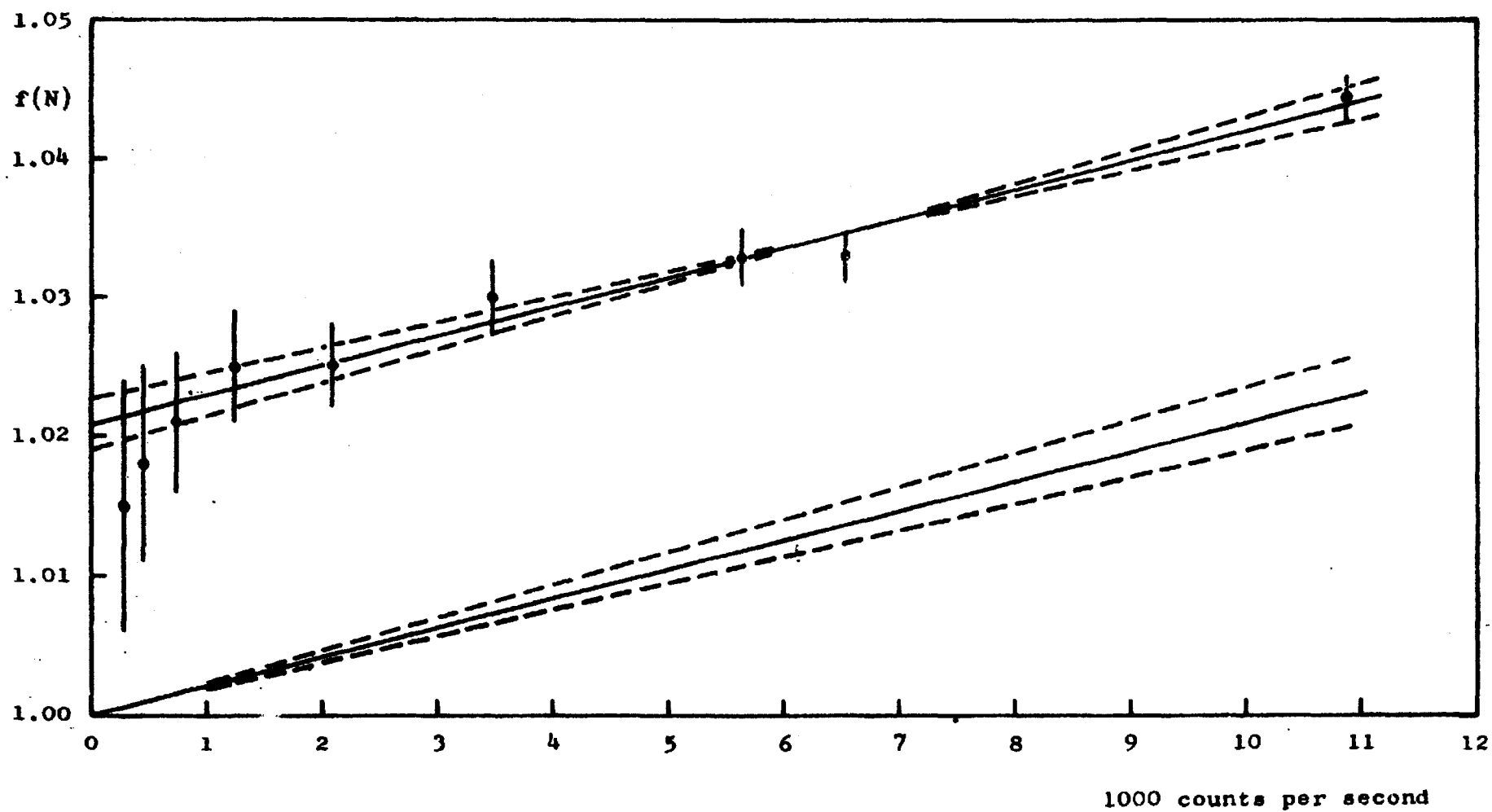


Fig. 6.1. Count-rate-dependent corrections.

$$\begin{aligned}
 A &= \frac{N_{\beta} \cdot N_{\gamma}}{N_c - N_r} \\
 &= \frac{N_{\beta} \cdot N_{\gamma}}{N_c \left(1 - \frac{N_r}{N_c}\right)} \\
 &\approx N \left\{ 1 + 2 \tau_c (1 - \epsilon_{\beta})(1 - \epsilon_{\gamma}) N \right\} ,
 \end{aligned} \tag{6.34}$$

and comparing this with (6.23), we find

$$\tau_c = \frac{1}{2(1 - \epsilon_{\beta})(1 - \epsilon_{\gamma})} . \tag{6.35}$$

In our measurements (table 6.1) we had $\epsilon_{\beta} = 0.634$ and $\epsilon_{\gamma} = 0.122$, and thus by (6.32) we find

$$\tau_c = 3.3 \pm 0.3 \text{ } \mu\text{s} , \tag{6.36}$$

which is in perfect agreement with the setting of the instrument. It is therefore reasonable to use formula (6.34) for this instrument when we are measuring gold foils of other thicknesses. Our measurements have further showed that the linear correction is valid up to 10 000 counts per second, and something serious happened at higher count-rates.

The Half-Life of the Foil Activity

Returning to the measurements given in table 6.1, we can now use the above results to correct the observed count-rates and then determine the half-life of the activity. Table 6.2 gives the thus corrected count-rates A_1' and A_2' and their logarithms. Since we are interested in a possible divergence from the half-life 2.7 days, we calculate the difference

$$\begin{aligned}
 (A') &= \log A' - \log e^{-\frac{\ln 2 t}{2.7}} \\
 &= \log A' + \frac{\log 2}{2.7} \cdot t ,
 \end{aligned} \tag{6.37}$$

Table 6.2

Ref. time 1965 24 Mar 10 ^h 15 ^m	A' ₁ cps	A' ₂ cps	$\frac{0.30103}{2.7} \cdot t$	log A' ₁	log A' ₂	$\Delta(A'_1)$	$\Delta(A'_2)$
+ 0.04167	6303 ± 8		0.00464	3.79955		3.8042 ± 0.0006	
+ 2	3820 ± 6		0.22298	3.58206		3.8050 ± 0.0007	
+ 4.04167	2262 ± 5		0.45062	3.35449		3.8051 ± 0.0009	
+ 4.10417	2223 ± 5	3470 ± 6	0.45759	3.34694	3.54033	3.8045 ± 0.0009	3.9979 ± 0.0007
+ 6	1363 ± 4	2136 ± 5	0.66896	3.13450	3.32960	3.8035 ± 0.001	3.9986 ± 0.0009
+ 8	817 ± 3	1277 ± 4	0.89194	2.91222	3.10619	3.8042 ± 0.0015	3.9981 ± 0.001
+ 10	489 ± 2	765 ± 3	1.11493	2.68931	2.88366	3.8042 ± 0.002	3.9986 ± 0.0015
+ 12	293 ± 1.6	454 ± 2	1.33792	2.46687	2.65706	3.8048 ± 0.002	3.9950 ± 0.002
+ 14	175 ± 1.3	273 ± 1.6	1.56090	2.4304	2.43616	3.8039 ± 0.003	3.9971 ± 0.002
+ 16	106 ± 1	164 ± 1.3	1.78389	2.02531	2.21484	3.8092 ± 0.004	3.9987 ± 0.003

where t is the time in days. Figure 6.2 shows $\Delta(A_1')$ and $\Delta(A_2')$, and the lines which have been drawn through the points are found as follows: We have for each foil a set of measurements of the form

$$\begin{aligned} (t_1, \Delta_1 \pm \sigma_1) \\ \vdots \\ (t_n, \Delta_n \pm \sigma_n) \end{aligned} \quad (6.38)$$

(table 6.2), and we want the probability distributions of the constants a and q in a line,

$$y = at + q, \quad (6.39)$$

"going through" all the points.

The probability that the line (6.39) goes through the point $(t_i, \Delta_i \pm \sigma_i)$ is then

$$\frac{1}{\sigma_i \sqrt{2\pi}} e^{-\frac{(y_i - \Delta_i)^2}{2\sigma_i^2}} \quad (6.40)$$

where

$$y_i = at_i + q, \quad (6.41)$$

and the probability that all n points lie on the line (6.39) is the product

$$\begin{aligned} \frac{1}{\sigma_1 \cdots \sigma_n} \left(\frac{1}{\sqrt{2\pi}} \right)^n e^{-\frac{1}{2} \left(\frac{(y_1 - \Delta_1)^2}{\sigma_1^2} + \cdots + \frac{(y_n - \Delta_n)^2}{\sigma_n^2} \right)} \\ = \frac{1}{\sigma_1 \cdots \sigma_n} \left(\frac{1}{\sqrt{2\pi}} \right)^n e^{-\frac{1}{2} f(a, q)}, \end{aligned} \quad (6.42)$$

where

$$f(a, q) = \frac{(t_1 a + q - \Delta_1)^2}{\sigma_1^2} + \cdots + \frac{(t_n a + q - \Delta_n)^2}{\sigma_n^2}. \quad (6.43)$$

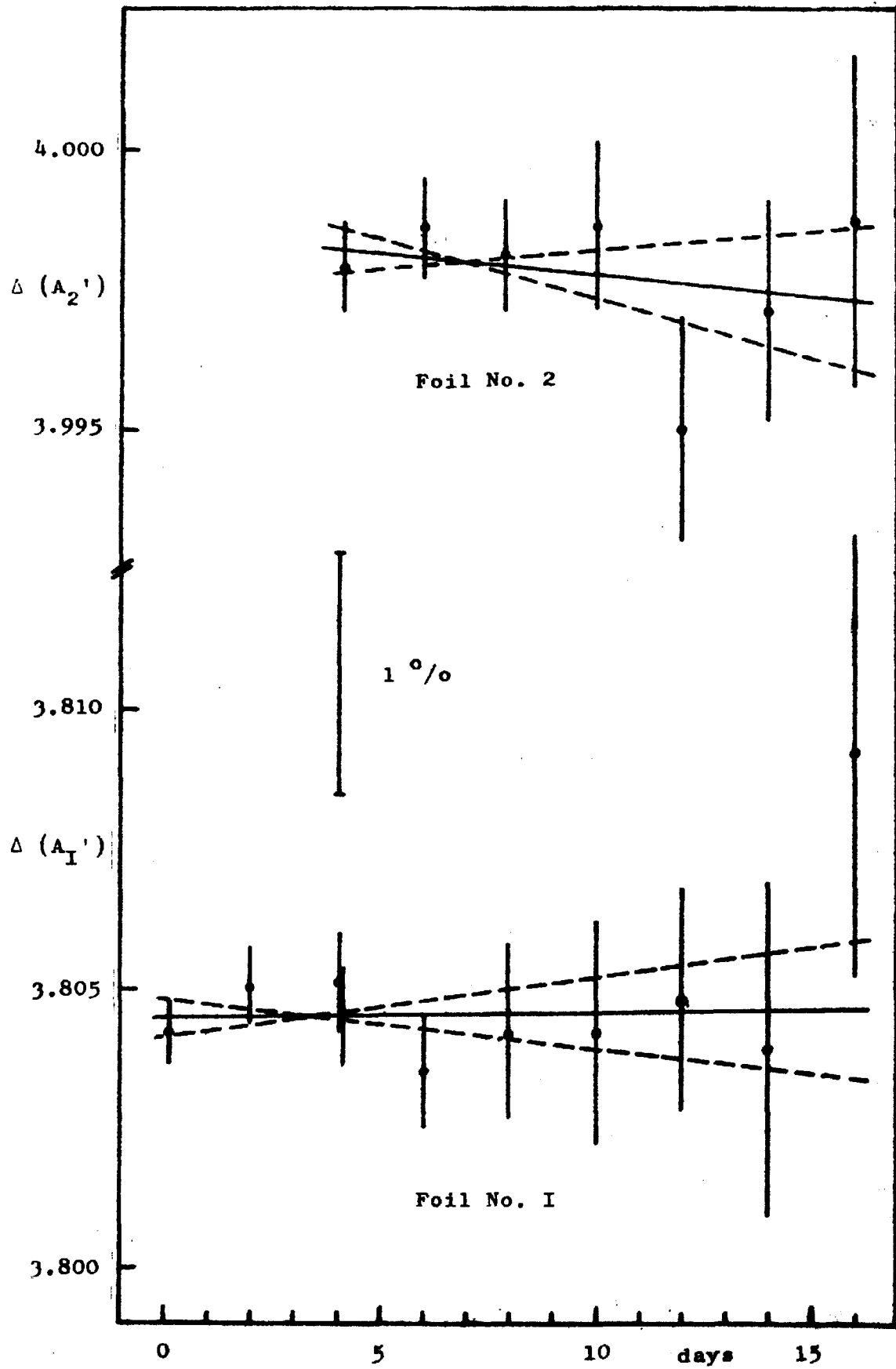


Fig. 6.2. Comparison with $T_{1/2} = 2.700$ days.

Since

$$(t_i a + q - \Delta_i)^2 = t_i^2 a^2 + q^2 + \Delta_i^2 + 2 t_i a q - 2 t_i \Delta_i a - 2 \Delta_i q , \quad (6.44)$$

we can rearrange $f(a, q)$ in the following way:

$$f(a, q) = A a^2 + B q^2 + 2 C a q - 2 D a - 2 E q + F , \quad (6.45)$$

where

$$A = \frac{t_1^2}{\sigma_1^2} + \dots + \frac{t_n^2}{\sigma_n^2} \quad (6.46)$$

$$B = \frac{1}{\sigma_1^2} + \dots + \frac{1}{\sigma_n^2} \quad (6.47)$$

$$C = \frac{t_1}{\sigma_1^2} + \dots + \frac{t_n}{\sigma_n^2} \quad (6.48)$$

$$D = \frac{t_1 \Delta_1}{\sigma_1^2} + \dots + \frac{t_n \Delta_n}{\sigma_n^2} \quad (6.49)$$

$$E = \frac{\Delta_1}{\sigma_1^2} + \dots + \frac{\Delta_n}{\sigma_n^2} \quad (6.50)$$

$$\text{and } F = \frac{\Delta_1^2}{\sigma_1^2} + \dots + \frac{\Delta_n^2}{\sigma_n^2} . \quad (6.51)$$

Further we find

$$\begin{aligned} f(a, q) &= B q^2 - 2(E - C a)q + A a^2 - 2 D a + F \\ &= B \left(q - \frac{E - C a}{B} \right)^2 + \left(A - \frac{C^2}{B} \right) a^2 - 2 \left(D - \frac{E C}{B} \right) a + F - \frac{E^2}{B} \\ &= \frac{\left(q - \frac{E - C a}{B} \right)^2}{\frac{1}{B}} + \frac{\left(a - \frac{D - E C / B}{A - C^2 / B} \right)^2}{\frac{1}{A - C^2 / B}} + \text{a constant} . \end{aligned} \quad (6.52)$$

After inserting (6.52) in (6.42) and integrating over q from $-\infty$ to $+\infty$ we find a Gaussian probability distribution for the slope a with the mean value

$$E[a] = \frac{D - EC/B}{A - C^2/B} \quad (6.53)$$

and the standard deviation

$$\sigma(a) = \sqrt{\frac{1}{A - C^2/B}} \quad (6.54)$$

(6.53) and (6.54) are identical with the formulae which can be derived by minimizing the sum of the squared differences as done in the method of least squares (e.g. A. Hald, 1952).

For every fixed value of a we have in the same way a Gaussian distribution in q with the mean value

$$E[q] = \frac{E}{B} - \frac{C}{B} a, \quad (6.55)$$

and the line (6.39) will thus for every a go through the point

$$(t_0, y_0) = \left(\frac{C}{B}, \frac{E}{B} \right). \quad (6.56)$$

From table 6.2 we thus find

$$E[a_1] = (0.16 \pm 1.00) \times 10^{-4} \quad (6.57)$$

and

$$E[a_2] = (-0.77 \pm 1.40) \times 10^{-4}, \quad (6.58)$$

and the mean of these two values is

$$E[a] = (-0.15 \pm 0.8) \times 10^{-4}. \quad (6.59)$$

This does not cause any change in the value 2.7 days used for the half-life since the correct half-life $T_{1/2}$ is found from

$$\frac{\log 2}{2.7} = \frac{\log 2}{T_{1/2}} - (0.15 \pm 0.8) \times 10^{-4}, \quad (6.60)$$

from which we find

$$\begin{aligned} T_{1/2} &= 2.7000 - 0.0004 \pm 0.0019 \\ &= 2.700 \pm 0.002 . \end{aligned} \quad (6.61)$$

This is in perfect agreement with the value

$$T_{1/2} = 2.697$$

recommended for use in the international comparison of measurements of Au^{198} (Comite consultatif pour les etalons de mesure des radiations ionisantes, 4^e session, annexe 4, 1963).

We will here just take this agreement as a proof of the radiochemical purity of the fold foils used.

CHAPTER 7:

CONCLUSION

One of the fundamental problems in the interpretation of experiments of a statistical nature, such as the observation of decays of radionucleides, is the inversion of probability theory as first treated in the famous article by the Reverend Thomas Bayes (1763). Since those days it has become more and more evident that the Bayes postulate on the "a priori" probability has to be generalized in order to cope with continuously variable parameters.

In ref. II and in more detail in ref. III a possible generalization to the continuous case is evaluated, and the criteria used in the generalization are pointed out to be based on a physical argument. The idea originates from the fact that the substitution method is the only primary method of establishing the unlinearity of a measuring instrument. The applicability of this method to nuclear counting instruments has been evaluated in ref. I.

In the present work the generalized Bayes postulate is used in the interpretation of coincidence experiments and thus provides a detailed understanding of the mathematical assumption necessary in order that the method may be used in a simple way for the determination of absolute disintegration rates. The original postulate by Bayes (1763) is derived as a special case of the generalized postulate.

In the ideal limit of the beta-gamma-coincidence experiment each decay can be detected in two independent ways by two detectors of different kinds, and the probability ϵ_c of simultaneous detection can be expressed by the detection probabilities ϵ_β and ϵ_γ of the two detectors respectively

$$\epsilon_c = \epsilon_\beta \cdot \epsilon_\gamma. \quad (7.1)$$

In chapter 4 it is demonstrated that (7.1) also holds as a relation between the mean values of the detection probabilities, where the means are taken over the whole volume of the radioactive source and over the observation time. It is here necessary to assume that the distribution of the activity in space and time can be expressed as a product in which the variables are separated in specified groups. From the analysis it is concluded that the ideal beta-gamma-coincidence experiment can be described by the mean value of the activity and the mean values of the two detection probabilities, and with the simple expression (7.1) for the mean probability of simultaneous (coincidence) detection.

In the actual experiment, the practical application of the results derived is demonstrated in the absolute measurements of neutron-induced radioactivity in gold foils. It is shown, partly by the design of the instrument and partly by specially arranged measurements, that the actual experiment is described by the theory. In the case of a 20 mg/cm^2 gold foil the accuracy is shown to be from 0.1 to 0.2 per cent; the 0.1 per cent is due to a correction for finite foil thickness arising mainly from an internal conversion in the dominating gamma ray following the Au^{198} decay (from the 411 keV level in Hg^{198}).

The most prominent class of corrections in coincidence counting is the count-rate-dependent corrections, including those for effects of dead time, accidental coincidences, piling up of electronic pulses, etc. It is here shown that even under the most simplifying assumptions the existence of a finite dead time will change the nature of the probability distributions on which the whole analysis is based. The derived interpretation of the measurements is thus only valid in the limit of low count-rates. The problem is solved by the substitution method, and as a special result the half-life of the activity in the gold foils is found with a relative standard deviation of 0.1 per cent. The half-life measured agrees with the value recommended for use in the international intercomparisons arranged by the Bureau International des Poids et Mesures, but since no other determinations have been made of the radiochemical purity of the gold foils used, it is necessary to

use this determination of the half-life as a proof that the gold foils are radiochemically pure.

In the present work the technical reason for measuring the activity of gold foils has been the wish to standardize the neutron density in the beam, which is used in an experiment for the determination of the neutron half-life (C. J. Christensen, A. Nielsen, A. Bahnsen, W. K. Brown, and B. M. Rustad, 1966). The standardization by the gold activation is performed simultaneously with a standardization by a completely independent method utilizing a He^3 proportional counter (J. Als-Nielsen, A. Bahnsen and W. K. Brown, 1966), which has also been developed to an accuracy of the order of 0.1 per cent. The determinations of the neutron density by these two methods agree within the calculated accuracy.

The generalized Bayes postulate and the use of the idea of inverse probability which it permits, are thus proved to be a powerful tool in the interpretation of physical experiments of this kind.

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ERRATA TO RISÖ REPORT NO. 70

P. 37: $E[a^2] = (n_1 - n_2)^2 + (n_1 + n_2 + 3) + (n_1 - n_2) \frac{Y}{X}$

should read

$$E[a^2] = (n_1 - n_2)^2 + (n_1 + n_2 + 2) + (n_1 - n_2) \frac{Y}{X} .$$

This error enters into the expression for $D^2[a]$, and thus 3 should be changed to 2

twice on page 24

once on page 25

twice on page 28

once on page 30

three times on page 37

and once on page 38

P. 60, ref. 3): Rev. Mod. Phys. 25, 818 (1957)

should read Rev. Mod. Phys. 25, 818 (1953).